Risk assessment of chronic arsenic exposure to human and domestic animals and possible remedial strategies: Study in selected endemic areas of West Bengal, India

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THESIS DETAILS

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Dedicated to

My parents

“Because of you, I’m here.”

STATEMENT OF ORIGINALITY

I, **Antara Das**, registered on **3rd January, 2019**, do hereby declare that this thesis entitled "**Risk assessment of chronic arsenic exposure to human and domestic animals and possible remedial strategies: Study in selected endemic areas of West Bengal, India**" contains literature survey and original research work done by the undersigned candidate as part of Doctoral studies.

All information in this thesis have been obtained and presented in accordance with existing academic rules and ethical conduct. I declare that, as required by these rules and conduct, I have fully cited and referred all materials and results that are not original to this work.

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This is to certify that the thesis entitled "**Risk assessment of chronic arsenic exposure to human and domestic animals and possible remedial strategies: Study in selected endemic areas of West Bengal, India**" submitted by **Ms. Antara Das** who got her name registered on **3rd Janaury, 2019** for the award of Ph.D. (Science) degree of Jadavpur University is absolutely based upon her own work under the supervision of **Dr. Tarit Roychowdhury** and that neither her thesis nor any part of the thesis has been submitted for any degree/diploma or any other academic award anywhere before.

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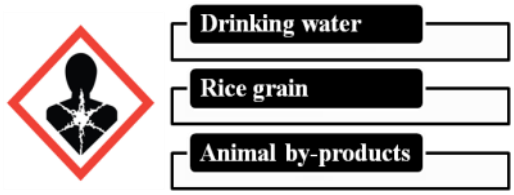
ABSTRACT



The present research is a multi-dimensional approach of the effect of arsenic (As) exposure in human, livestock and environment. It is a naturally occurring toxic metalloid, globally classified as a group I human carcinogen. Groundwater As contamination is a natural calamity exhumed from its sediments and ruling the world's largest delta Ganga-Meghna-Brahmaputra for many years. Contamination of As is not only limited to groundwater, with time it has spread into the food chain. Nine districts of West Bengal, India, covering~ 6 million people are severely suffering from groundwater As contamination. In many of the remote villages of West Bengal, As kills people slowly and silently. The work firstly covers the evaluation of 20 physico-chemical parameters in 110 groundwater samples from all the blocks of Nadia district located in West Bengal, for its overall quality assessment. A Water Quality Index (WQI) modelling has been done further where ~ 66% of water sample are not recommended for use. The mean anionic concentration range in groundwater seems to be in the order of bicarbonate (HCO_3^-)>chloride (Cl^-)>carbonate (CO_3^{2-})>sulphate (SO_4^{2-})>nitrate (NO_3^-)>phosphate (PO_4^{3-}) signifying the water quality of Nadia is alkaline, mainly due toHCO_3^- alkalinity. Hardness is determined by Ca^{2+} and Mg^{2+} ions where Ca^{2+} ion concentration (mean: 53.7 mg/L, range: 4–156 mg/L) overrules Mg^{2+} one (mean: 44.9 mg/L, range: 0.18–114 mg/L). Groundwater in all the blocks is As contaminated and maximum As concentration is 206 µg/L in Chakdah. Few ground water samples have been identified with elevated NO_3^- (45–57.6 mg/L), particularly from four blocks (Krishnanagar-I, Nabadwip, Kaliganj and Chapra) in the district. No sample has been identified with uranium (U) concentration (range: 0.21–20.9 µg/L) beyond its permissible limit (30 µg/L, recommended by WHO). Presence of high Cl^-, NO_3^- and

especially HCO_3^- ions in groundwater and their positive interdependence among each other may magnify U contamination through dissolution of U as they are good carrier of U. The F^- concentration has been observed within its recommended value in drinking water. There is a strong possibility for cancer risk through As and U in drinking water as the mean cancer risk value (1.2×10^{-3} and 2.48×10^{-3}) goes beyond their respective acceptance level. In few cases NO_3^- poses non carcinogenic health hazards in future. Secondly, As contamination scenario in groundwater and treated drinking water sources from all the 9 gram panchayats under Raninagar-II block, Murshidabad has been studied. Around 54.6% of groundwater samples from the domestic and community tube-wells (n = 366) are with As concentration above the permissible limit in drinking water (10 μg/L); while 37.3 % of groundwater samples from agricultural tube-wells (n = 67) are As-contaminated above its recommendation in irrigational water (100 μg/L). About 12.5 tonnes of As is withdrawn annually through irrigational water in the entire block. Bio-accumulation of As in rice grain (1.78) shows the potential of hyper-accumulation. The concentrations of two essential micro-nutrients in groundwater (Selenium and Zinc) are low (mean: 0.57 and 84.5 μg/L) which encourages the poor health of the local inhabitants. The populations suffer from serious cancer and non-cancer risk through contaminated drinking water where the studied adult males face higher risk than the females. Hence, the role of the key factors (As intake, age and sex) regulating As toxicity is aimed to evaluate in a severely exposed population from Murshidabad. The mean As concentrations in drinking water through tube-well, Sajaldhara treatment plant and pipeline were observed as 208, 27 and 54 μg/l, respectively. Urinary As concentrations were found as < 3–42.1, < 3–56.2 and < 3–80 μg/l in children, teenagers and adults, respectively. The mean As concentrations in hair and nail were found to be 0.84 and 2.38 mg/kg; 3.07 and 6.18 mg/kg; and 4.41 and 9.07 mg/kg, respectively, for the studied age-groups. Arsenic concentration in drinking water was appeared to be associated with that in hair and nail

more than urine. Deposition of As in biomarkers appeared to be dependent on age; however, independent of sex. A principal component analysis showed a direct relationship between dietary intake of As and deposition in chronic biomarkers. Nail was proved the most fitted biomarker of As toxicity by Dunn's post hoc test. Monte Carlo sensitivity analysis and cluster analysis showed that the most significant factor regulating health risk is 'concentration of As' than 'exposure duration', 'body weight' and 'intake rate'. The contribution of As concentration towards calculated health risk was highest in teenagers (45.5–61.2%), followed by adults (47.8–49%) and children (21–27.6%). Besides sub-clinical toxicity, different skin lesions were observed among the affected inhabitants like 'raindrop pigmentation' in the dorsal part of the hand, feet or whole body, 'spotted and diffused keratosis', 'melanosis', even 'bowens'. Arsenic toxicity in the domestic livestock and possible health risk for human and environment caused by them is also assessed. Daily dietary As intake of an exposed adult cow or bull is ~ 4.56 times higher than control populace. Arsenic toxicity is well exhibited in all the biomarkers through different statistical interpretations. Arsenic bio-concentration is faster through water compared to paddy straw and mostly manifested in faeces and tail hair. Cow dung and tail hair are the most pronounced pathways of As biotransformation into environment. A considerable amount of As has been observed in animal proteins, such as cow milk, boiled egg yolk, albumen, liver and meat from the exposed livestock. Cow milk As is mostly accumulated in casein due to the presence of phosphoserine. SAMOE–risk thermometer, calculated for the most regularly consumed foodstuffs in the area, shows the human health risk in an order of: drinking water > rice grain > cow milk > chicken > egg > mutton. USEPA health risk assessment model reveals more risk in adults than in children from the foodstuffs where the edible animal proteins cannot be ignored. This research focused on investigation of the performance efficiencies of the mitigation measures made by the government and non-governmental organizations. It observed that in some areas, the pipeline supplied

water and the treated water from the arsenic-iron removal plants (AIRP) contain adequate arsenic to cause health risk. The present work found annual average As removal efficiency of the studied 12 AIRPs in Gaighata, North 24 Parganas is 61.2% (range: 35.2 to 82.6%) and the annual average Fe removal efficiency of the AIRPs is 81.4% (range: 35.7 to 97.3%). Trace element study shows presence of Al and Mn in 30 and 50 % treated samples respectively. WQI study revealed that 25% treated water samples are of 'poor' quality; 16.7% treated water samples are of 'high' heavy metal evaluation index value. Therefore, besides insufficient access to safe water, in many areas, people still drink contaminated water in the name of 'treated water'. Even in my survey period of one year, two plants also got closed due to lack of manpower or poor performance which reveals that the plants are not maintained properly and regularly. This depicted the failed scenario of the As mitigation plans by the concerned authorities. The Reverse Osmosis (R.O.) and Sajaldhara water treatment plants in Raninagar II, Murshidabad showed 77.6 and 74.4 % of As removal efficiency. Among all kinds of alternate drinking water sources, dug wells seemed to be the safest in regards to As contamination with a natural Fe/As ratio of 66 and account for lowest health risk as per Severity Adjusted Margin of Exposure. Although, certain percentage of dug well water quality is hard, that may be removed through boiling. Lastly, a solar oxidation process is promoted through my research for removal of As from contaminated water (removal efficiency ~ 50%) with application of amla at minor dose. The suggestion is to increase the use of dug wells in the villages at both domestic and community level along with the usage of surface water bodies. Domestic livestock too should be fed with surface water instead of groundwater. Furthermore, prohibition in exploitation of groundwater is the utmost call of this hour to build a sustainable future on this earth.

HIGHLIGHTS

- After several mitigation strategies undertaken, groundwater As contamination still persists
- Pipeline supplied water and treated water from As removal plants are not at all safe
- Nearly 66% of groundwater of Nadia district is not recommended for domestic/drinking purpose
- All the 17 blocks of Nadia are As-contaminated with highest concentration of 206 μg/L
- High levels of Cl^{-}, NO_3^{-} and HCO_3^{-} ions in groundwater of Nadia may magnify U contamination in future
- Population of Nadia suffer from substantial high cancer risk through As and U (mean: 1.2×10^{-3} and 2.48×10^{-3})
- About 54.6 % of domestic and community tube-wells in Raninagar II, Murshidabad contain As >10 μg/L
- Withdrawal of As in Raninagar II through irrigational water is 12.56 tonnes/year; deposition rate in paddy fields is 1.08 kg/ha
- Rice grain is a potential hyper-accumulator of As
- Deposition of As in human biomarkers depends on age; independent of sex
- Nail As concentration is proved the most appropriate biomarker of evaluating As toxicity
- The most significant factor that regulates health risk is ‘concentration of As’ than ‘exposure duration’, ‘body weight’ and ‘intake rate’
- Domestic livestock are exposed to As toxicity as well as humans
- Dietary intake of As per day is ~4.5 times higher in cattle of As exposed areas than control
- Animal proteins contain As; cow milk causes highest health risk followed by chicken, egg and goat flesh.
- In whole milk, casein accumulates maximum As (83%) due to presence of phosphoserine
- Arsenic is bio-transferred from livestock to the environment by their excreta
- Arsenic gets biomagnified in humans through consumable animal proteins along with As-contaminated water and rice
- Among all kind of available drinking water sources, dug wells seem to be the safest one with lowest health risk
- Solar Oxidation process can be promoted at domestic level for de-arsenification of contaminated drinking water with application of small doses of amla.

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Chapter 1:

Introduction

1.1. Groundwater resources in India

Survival of human beings immensely depends on freshwater, which is required for drinking purposes, agricultural activities or other domestic tasks. Around 3% of the total water in the world is fresh water. India owns only about 4% of the world's freshwater resources, while it is still the leading country in groundwater usage (World Bank, 2012). The demand of freshwater in India is increasing rapidly with a speedy growth in population, so, the amount of groundwater is in a perilous condition now. India, the largest user of groundwater resources in the realm where ~ 90% of groundwater is used for irrigation and the remaining 24 billion cubic meters fulfills about 85% of the country's drinking water requirements (India News, 2022). Due to misuse of groundwater, numerous aquifers are unfeasible now and India is turned into a 'water stressed region' with a per capita utilizable amount of 1122 m^3/year. It is anticipated that by the end of 2050, India will be on the edge of running out of water, when the population is expected to become stable (Gupta and Deshpande, 2004). According to the 2017 CGW (Central Groundwater Board) report, ~ 40% of the 700 Indian districts reportedto have groundwater levels termed as 'critical' or 'overexploited'. On the word of "NITI Aayog Report" of 2019, the water demand of India will be twice of the available resource by the year 2030 (The Statesman, October, 2022).

1.2. Groundwater quality in India

Almost 80% of illnesses in human beings are initiated by contaminated water, so, the status of groundwater is obviously a major matter of concern (Ramakrishnaiah et al., 2009; UNESCO, 2007). Beyond 33% of groundwater resources in India is unsafe for consumption (Chakraborti et al., 2010; Times of India, 2010). Several unpleasant toxic contaminants are polluting groundwater frequently from different sources whether through natural, agricultural or industrialized activities, that origins a major intimidation to health (Wu and Sun, 2016). Moreover, the daily and excessive exploitation of groundwater catalyze the chemical weathering of minerals beneath the aquifers under different hydrogeochemical environments that actually kicks off natural groundwater contamination. The major groundwater contaminants are fluoride (F^-), iron (Fe), arsenic (As), nitrate (NO_3^-), and uranium (U). The movement of water and dispersal within the aquifer spreads the pollutants over a broader area.

1.2.1. Groundwater contamination through fluoride, nitrate and uranium

Fluoride causing diseases have spread in nearly 40% of theIndian population particularly South India (Chakraborti et al., 2016). Raj and Shaji (2017) and Singaraja et al. (2013) elaborated the picture of sufferings due to F^- pollutionin groundwater. In this state, Bankura, Puruliya and Birbhum are also acknowledged with groundwater F^- pollution as reported in Batabyal and Gupta (2017); Datta et al. (2014); Mondal and Kumar (2017), Mondal and Nath (2015) and Samal et al. (2015). Groundwater F^- contamination is mostlycaused by the solubility of minerals e.g. fluorite (CaF_2), hornblende [$NaCa_2(Mg,Fe,Al)_5(Si,Al)_8O_{22}(OH,F)_2$] or fluorapatite [$Ca_5(PO_4)_3(Cl,F,OH)$] (Narsimha and Rajitha, 2018). Fluoride burden in the adjacent aquifers is also ruled by different metamorphic or sedimentary rocks and their degree of fluorine enrichment (Routroy et al., 2013). Again, groundwater of India is contaminated with NO_3^-; the states are Uttar Pradesh (Agarwal et al., 2019; Sankararamakrishnan et al., 2008), Andhra Pradesh (Adimalla et al., 2018; Rao et al., 2017), and Rajasthan (Jandu et al., 2021; Rahman et al., 2021). In the year 2008, the district Hooghly in West Bengal was recognized with NO_3^- amelioration in its groundwater and surface water, as reported by Kundu et al. (2008). Nitrate contamination is typically an output of agronomic activities with superfluous usage of nitrogen-bearing fertilizers. As nitrate is greatly soluble in water, it percolates in groundwater during moving through the sub-soil layers, with low little NO_3^- retention ability of the soil particles (Majumdar and Gupta, 2000). Another accountable mineral for groundwater impurity is U that causes a major health risk having severe radiological and chemical effects and nephrotoxic adversities (Kumar et al., 2011; Singh et al., 2013). Generally, the iron hydroxides (ferrihydrite) and amorphous iron oxide are decent sink of U (Das et al., 2018; Fendorf, 2016). Besides, mine leaching causes natural U moveinto groundwater from its diverse mineralized forms: pitchblende (U_3O_8), uraninite (UO_2), uranyl nitrate ($UO_2(NO_3)_2$), uranophane ($Ca\ (UO_2)_2–SiO_3(OH)_2{\cdot}5H_2O$), autunite ($Ca(UO_2)_2(PO_4)_2.10–12H_2O$), etc. (Wu et al., 2014). Henceforth, the groundwater quality is required to be monitored regularly for wide use by the population.

1.2.2. Groundwater contamination through arsenic

Groundwater As contamination is a natural menace distressing countries all over the globe, especially the southeastern countries such as India, Bangladesh, China, Vietnam (Ravenscroft et al., 2011). Worldwide, groundwater As is majorly associated to

degradation of sediments and rocks, desorption from As enriched pyrites and hydroxides, rainwater interceded deposits of weathered sulphide, which are further controlled by pH, dissolved organic carbon (DOC), redox chemistry and several anions (Alexakis et al., 2021). Until now, groundwater As contamination has affected 20 states and 4 UT (Union Territories) in India (Shaji et al., 2021). Above 500 million populaces in the Ganga–Meghna–Brahmaputra (GMB) delta, covering~ 569,749 km^2 area suffer from As contamination (Chakraborti et al., 2004, 2013). Near 359 million people from India live in As endemic states where approximate 70.4 million people from 6 states are at risk of groundwater As toxicity (Chakraborty et al., 2016). States like West Bengal, Bihar, Chattisgarh, Uttar Pradesh, Jharkhand, Assam, Manipur are known with groundwater As contamination since past 30 years (Chakraborti et al., 2018; Das et al., 2020; Rahman et al., 2009). In West Bengal, nine districts covering ~ 6 million population undergo high risk due to As contaminated drinking water (Chakraborti et al., 2015; Dey et al., 2014; Rahaman et al., 2013; Santra, 2017). The district Nadia at the east bank of the river Bhagirathi in West Bengal is extremely polluted with As, reported earlier (Das et al., 1996; Mazumder et al., 2010; Rahman et al., 2014; Roychowdhury et al., 2008). Rahman et al. (2014) conveyed that groundwater in Nadia is affected with As; near about 51.4% of the tube-wells had As beyond 10 μg/L. Twenty-five, out of a total number of 26 blocks in Murshidabad district were found to be As-affected where As concentration in groundwater ranged 3–3000 μg/L with a mean concentration of 240 μg/L (Chakraborti et al., 2009; Das, 2013). Lalgola, Raghunathganj-II, Bhagwangola-I & II, Raninagar-I & II, Berhampore, Hariharpara, Domkal, Jalangi are some of the worst As-affected blocks. Rahman et al. (2005a) reported that approximately 53.8% of the groundwater samples (n = 29,612) analyzed from the district in the depth of 10–120 m contained As beyond its acceptable limit in drinking water (10 μg/L).

1.2.2.1. History of arsenic

Arsenic is a natural contaminant in groundwater silently causing fatal troubles in human life. It is omnipresent in the environs and 20^{th} most copious element in the earth-crust. Though its compounds were excavated by the early Chinese, Greek and Egyptian civilizations, it is purported that As itself was first recognized by Albertus Magnus, a German alchemist, in 1250. It was considered as the "king of poisons" from the time of the Roman Empire. The world history remembers it as a ‘secret weapon’ being used in a series of minor doses to poison emperors, kings, or cardinals that produced loss of strength,

confusion and paralysis followed by death. Symptoms of As poisoning were challenging to identify, since they could imitate food poisoning and other conjoint disorders. The great revolutionary French leader Napoleon Bonaparte or the famous American politician Claire Booth Luce, Tiberius Britannicus, the son of Roman emperor Nero Claudius Drusus were the fatal victims of As poisoning. On the other hand, history also recalls As as the "Saviour of Syphilis" (Frith, 2013). In medical history, it was used to treat trypanosomiasis or sleeping sickness, and syphilis or great pox. Hippocrates used the As-sulphide mineral 'realgar' and 'orpiment' for the treatment of ulcers and abscesses. Also, several studies from China revealed that arsenic trioxide (As_2O_3) was prolific in treatment of de novo and relapsed acute promyelocytic leukaemia in the early 1990s. In Victorian times, Dr. Fowler's Solution (dissolved potassium arsenate) was a popular tonic used by Charles Dickens. Thereafter, organoarsenic compounds (roxarsone) were added to poultry feed to promote growth, improve weight gain and prevent disease. Organoarsenic compounds like roxarsone and and p-arsanilic acid are broadly used in poultry production as feed additives to control parasites, improve feeding practice, growth augmentation, and to progress the color of meat (Mangalgiri et al., 2015). Roxarsone is not freely adsorbed or metabolized, thus its concentration was documented in animal manures up to 40 mg/kg (Kyakuwaire et al., 2019). Due to environmental and human health concerns, arsenical compounds were dropped as feed additives by the European Union (EU) in 1999 in U.S. poultry production, some other countries including India still use them. Moreover, As is used as a doping agent in semiconductors for solid-state devices, and in bronzing, pyrotechnics and for hardening shot.

1.2.2.2. Characteristics of arsenic

Arsenic is ranked as a group I cancer-causing agent (carcinogen) according to the combination of its toxicity and potential for humanhealth risk by the IARC Monographs as well as ATSDR (Agency for Toxic Substances and Disease Registry) and USEPA (United State Environmental Protection Agency). It is colorless and odorless metalloid, with atomic number 33, atomic weight 74.9, density 5.73 g/cc and exists in the periodic table in group 15, between phosphorus and antimony ([Ar] $3d^{10}4s^{2}4p^{3}$) (**Fig. 1**). Elemental As is insoluble in water. It exists in four oxidation states i.e. -3, 0, +3 and +5. Arsenic is commonly present in inorganic states as oxyanions of trivalent arsenite (As-III) and pentavalent arsenate (As-V). Under oxidizing situations, arsenate is prevalent whereas, under anaerobic reducing conditions, it is more likely to be present as arsenite (IARC,

2012). Arsenic has an analogous ionization energy and electro negativity to its lighter congener phosphorus, therefore freely formulates covalent molecules with most of the non-metals. It naturally exists in an extensive range of minerals in soil and sediments in forms of inorganic compounds; metals of sulfides and oxide ores. The most common As containingminerals are arsenopyrite (FeAsS), realgar (AsS), arsenolite (As_2O_3), lollingite ($FeAs_2$, $FeAs_3$, $FeAs_5$) and orpiment (As_2S_3). Arsenic makes hygroscopic crystalline oxides (As_2O_3 and As_2O_5), which are colorless, odorless and readily solvable in water to form acidic solutions. Contamination in groundwater by arsenic mostly comes through arsenic (V) acid (weak acid) and its salts (arsenates). Being redox and pH sensitive component, its presence, distribution and mobility depends on pH, Eh and microbial activity. It is a strong neurological and liver toxin as well as a lung, bladder and skin carcinogen. Compounds of inorganic As are more toxic than organic ones, but they are transformed to inorganic compounds when absorbed in biological systems (Hughes et al., 2011). According to Roy et al. (2013), arsenites (As^{3+}) and arsenates (As^{5+}) are most frequentin the environment and the organic forms of Ase.g. monomethylarsenic acid (MMA), dimethylarsenic acid (DMA), trimethylarsine oxide (TMAO) and arsenobetaine are most common in the marine environment.

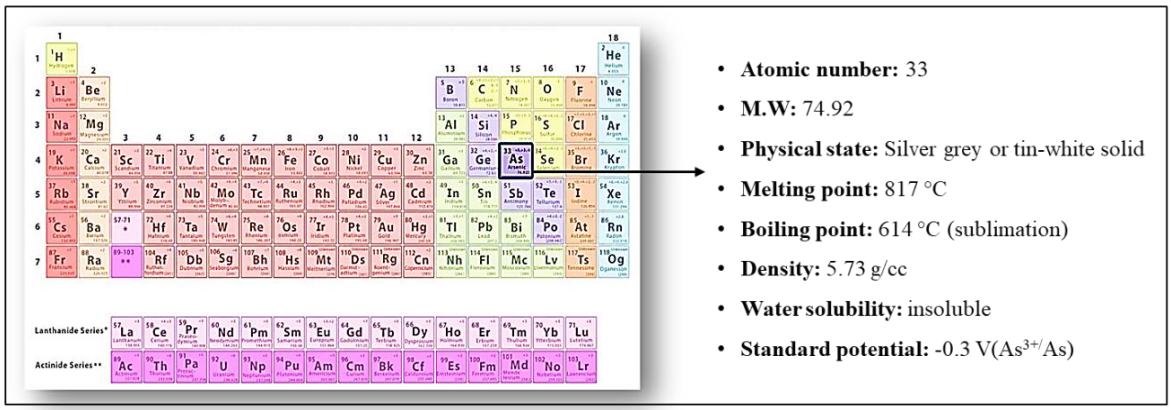


Fig. 1. Properties of arsenic

1.2.2.3. Source, mobility and distribution of natural arsenic

Groundwater As is generally responsible for the native geo-chemical atmosphereof the aquifers and adjacent soil. Depending upon several hydro-geochemical circumstances, arsenicarises into groundwater naturally through ‘oxidation of As containing minerals’ or ‘reductive dissolution of iron oxyhydroxide’ (Nickson et al., 2000). Groundwater As contamination is such a natural calamity exhumed from its sediments and ruling the

world's largest delta Ganga-Meghna-Brahmaputra delta for past 30–40 years (Chakraborti et al., 2013). West Bengal falls under the lower GMB basin which is loaded with As and the soil is largely alluvial (Chakraborti et al., 2013; 2017). The release and dispersal of As in the aquifers is generally controlled by quite a few factors like competitive ion exchange, pyrite oxidation and most significantly redox disequilibria (Chakraborty et al., 2015). Mobilization of As through the microbial breakdown of FeOOH under reducing environment is the most acknowledged hypothesis which is again supported by some strong correlations between dissolved As with Fe, methane, and ammonia found in groundwater (Chakraborty et al., 2015; Dowling et al., 2002; Kar et al., 2010). Also, some researches earlier discussed As mobilization through sulfide oxidation reactions during massive irrigation in dry summer (Chowdhury et al., 1999). Arsenic mobilization in alluvial delta is caused by inflow of detrital organic carbon through many biogeochemical processes too (Harvey et al., 2002).

1.3. Arsenic in food chain

Arsenic pollutes the total crop farming structure by tainted irrigational water (Allevato et al., 2019; Chowdhury et al., 2018a). A huge population suffers from its exposure through contaminated foodstuffs because cultivation with As contaminated irrigational groundwater compels the toxicant entering into the grains and vegetables where effect of contaminated soil is also significant (Roychowdhury et al., 2002a, b, 2005; Sanyal et al., 2002). Studies displayed that irrigational water contains enormous As concentration in the paddy fields of West Bengal; (<3 to 990 μg/L) in Raninagar-II, Murshidabad, (171 to 493 μg/L) in Gaighata, (74 to 301 μg/L) in Deganga, North 24 Paraganas etc. (Chowdhury et al., 2018a; 2020a; Das et al., 2021). Surface soil As concentrations were observed as 9.5 to 19.4 mg/kg in paddy fields of Murshidabad and 22.4 to 60.2 mg/kg in paddy fields of North 24 Parganas (Roychowdhury et al., 2005; Chowdhury et al., 2018). Accumulation of As in various parts of paddy plant has been researched previously (Rahman et al., 2007; Tuli et al., 2010). The studies showed As concentration is highest in rice hull followed by bran-polish, brown rice, raw rice and polished rice for both parboiled and non-parboiled varieties of rice. Arsenic gets accumulated and translocated from root, stem and finally reaches leaf and the grains (Bhattacharya et al., 2009, 2010). Prolonged As exposure through the consumption of contaminated crops and vegetables degrades health quality in various parts of the state (Bhattacharya et al., 2010; Santra et al., 2013). One of the prime sources of As exposure in humanin recent times is rice and rice-based products whereas

As concentrations in other pulses, cereals, vegetables also contribute to human health risk when consumed at a daily rate (Biswas et al., 2012, Upadhyay et al., 2019a, b). Also, as a significant cultivable crop, rice grain'sAs bio-accumulation showed the ability of hyper-accumulation (Samal et al., 2021). Thus, in addition to theingestion of drinking water, As contained foodstuffs, specially rice grains, escalate people's health risk (Ahmed et al., 2016; Joardar et al., 2021a,b).

Rice, the basic diet in most of the state, contributes daily a large quantity of As and stretches a substantial health risk to both As exposed and unexposed areas population (Biswas et al., 2019; Chowdhury et al., 2020a; Joardar et al., 2021a). Cultivation season, geographical identity, background water and soil characteristics, rice cultivars etc. forces rice grain As concentration to vary (Chowdhury et al., 2018a; 2020a). A highest 200 µg/kg As concentration has been advised for i-As in polished rice as per World Health Organization (WHO) and the Codex Alimentarius, (Codex Alimentarius Commission, 2014). Though, Meharg et al. (2006) stated that rice with i-As concentration over 100 µg/kg starts to enforce cancer risk in As affected countries. Parboiling of sunned rice with As tainted water or cooking of rice grain with As contaminated water creates an additional burden (Chowdhury et al., 2018b, 2020a).

1.4. Toxicity of arsenic and its mechanism in human

Organic As is less toxic than inorganic As (As-V and As-III). Inside human body, metabolism turns inorganic As into organic one that includes reduction of arsenate to arsenite, then methylation into monomethylarsonic acid (MMA) and dimethylarsinic acid (DMA) (Vahter, 1999). A higher i-As concentration decreases the methylation aptitude of As that results in higher inorganic urinary As (Torres-Sánchez et al., 2016). Human cells use phosphate for energy generation but arsenate can mimic and substitute the phosphate that impairs the ability of the cell of energy generation and interconnect with other cells. Metabolism of As is catalyzed by arsenite methyltransferase enzyme, a consecutive process of reduction to trivalent As from pentavalent As followed by oxidative methylation back to pentavalent state. However, arsine gas is the most toxic form of As; inhalation of over 10 ppm of which is fatal. Different forms of As and its methylation in human system is described in **Fig. 2**.

Among the inorganic forms, asenite is generally more toxic than arsenate. Acute effects of As cause gastrointestinal distress to death and chronic As exposure affect several major

organs in mammals. It causes malignant disruptions in skin and may also inhibit the proper functioning of the immune system, leading to viral/bacterial infections (Duker et al., 2005). In human systems, the mechanism of As in terms of toxicity is still the subject of research. Arsenite binds to sulfhydryl and disulfide groups of enzymes and thus disrupts their work; inhibits pyruvate and succinate oxidation pathways and the tricarboxylic acid cycle, triggering impaired gluconeogenesis and reduces oxidative phosphorylation. It targets omnipresent enzyme reactions, so disturbs almost all organ systems. Whereas, arsenate replaces the stable phosphates which leads to rapid hydrolysis of ATP formation. It initiates loss of high energy phosphate bonds and uncouples oxidative phosphorylation.

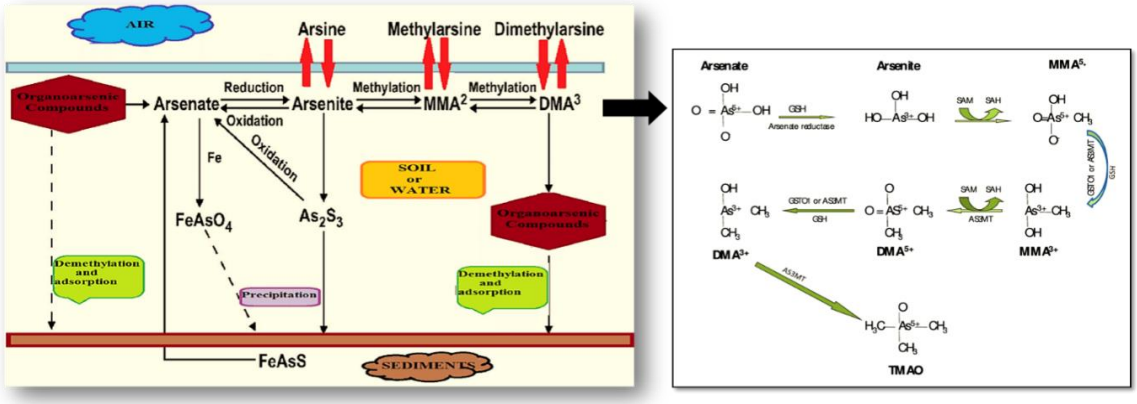


Fig. 2. Different forms of arsenic in biosphere and its methylation in human system (Navin et al., 2013**)**

However, it is reported that inorganic As is more toxic than its other metabolites; methylarsonic acid (MMA), dimethylarsinic acid (DMA), and the methylated forms of As are more readily excreted than inorganic As (Concha et al., 1998). The metabolism system results in elimination of half of ingested As in the urine in 3–5 days. DMA is the dominant urinary metabolite (60–70%) in comparison to MMA while a small amount of inorganic As is also excreted directly (Hopenhaynrich et al., 1993; Ratnaike, 2003). Hall (2002) says that around 60 to 90% of soluble inorganic As is absorbed via the gastrointestinal tract. About 50 to 70% of absorbed As is eradicated through kidneys by methylation and the rest accumulates in hair, nail, and other tissues (Nielsen, 2001). While urinary As is a good biomarker for recent exposure (Nermell et al., 2008; Vahter, 1994), evaluation of As in hair and nail tissues is the best way to measure chronic As exposure (Brima et al., 2006; Rahman et al., 2001; Samanta et al., 2004). Considerably greater amounts of As is accumulated in hair tissues than other biological samples due to keratin proteins which

constitute sulfur-containing amino acids (Byrne et al., 2010; Gault et al., 2008). Therefore, in children where skin manifestation from As toxicity is obscure, sub-acute or sub-clinical toxicity is assessed through the biomarkers. Different adverse impacts caused by As is depicted in **Fig. 3**.

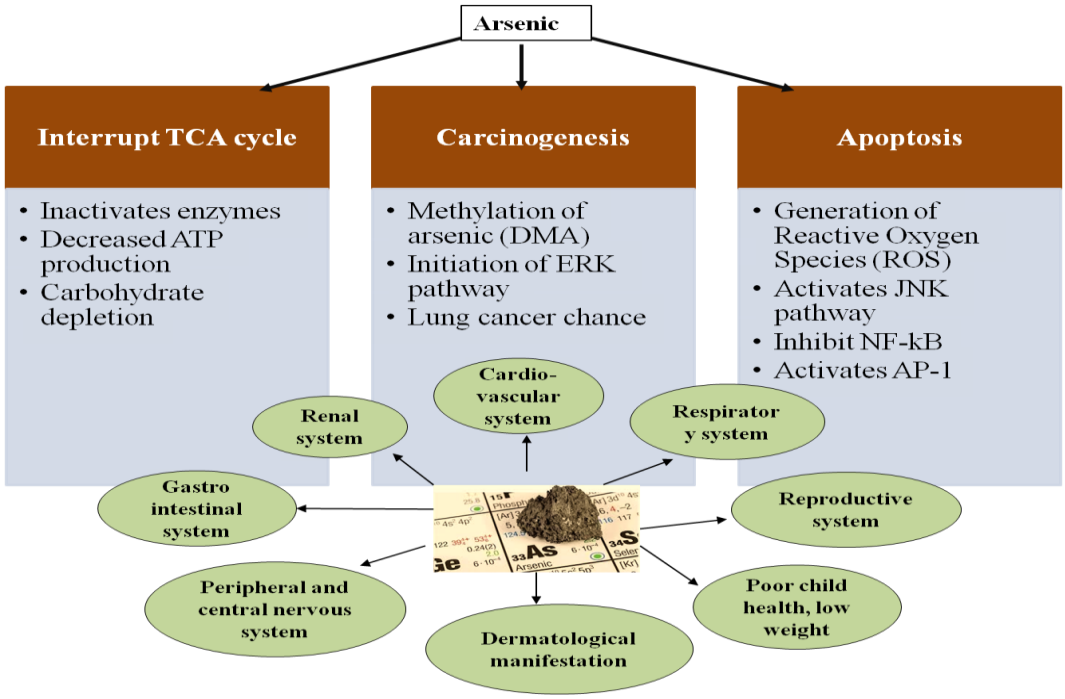
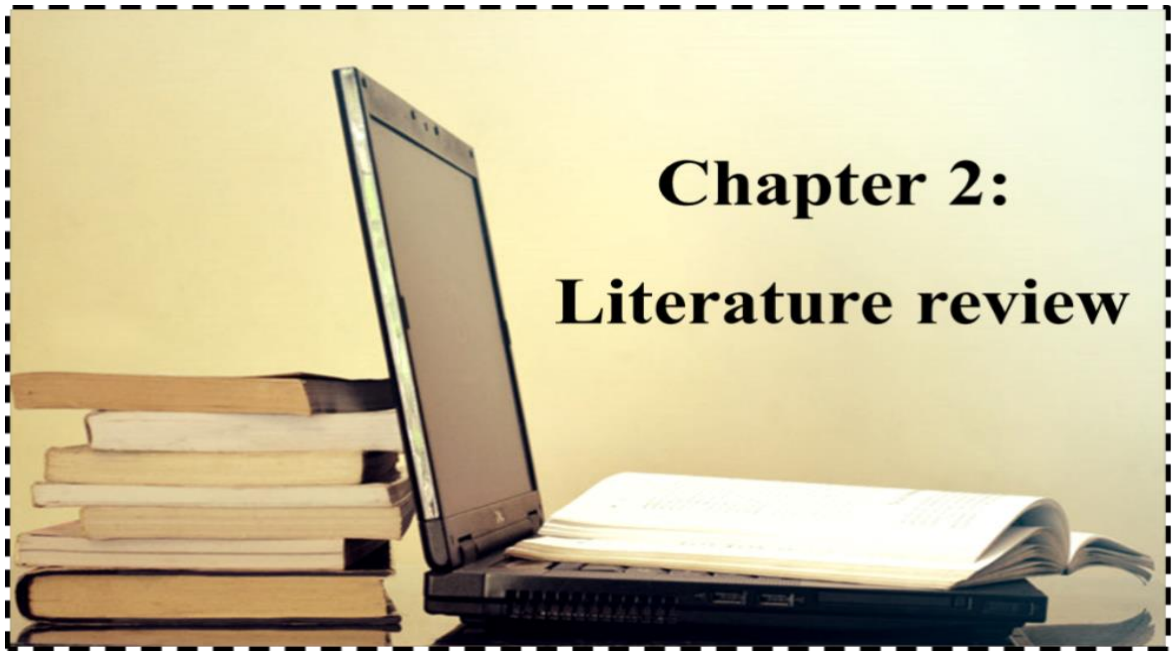


Fig. 3. Health effects of arsenic adversity

1.5. Arsenic toxicity in livestock (domestic and farm)

Arsenic accumulation in livestock generally don't show any significant skin manifestations but suffers from subclinical toxicity as it stays in their blood, keratin tissues and faeces (Mandal, 2017). It produces chronic effect in their body system such as loss of weight, low immunity, abdominal discomfort, diarrhea, low production efficiency, reduced milk yield and other diseases (Mandal, 2017, Roy et al., 2013). In the worst scenario, there are incidents of animal death in South America (Faires, 2004) and South Dakota (Neiger et al., 2004) suffering from high As toxicity. The symptoms were mostly ataxia, anorexia, diarrhea and lethargy. The dead calves' kidney As concentration was found to be 25-44 mg/kg. The soil, drinking water, green food with which they were raised up, contained As concentration 4.4 mg/kg, 0.027 mg/L and 360,000 mg/kg respectively (Neiger et al., 2004). Arsenic contaminated milk is secreted from cows who consume As contaminated water and paddy straws for long (Abedin et al., 2002). Eggs of laying hens in an Asexposed area of Bangladesh is reported with As concentration 19.2 µg/kg. With

increasing age of the hen, As content in eggs appeared to be increased by 0.94% per week (Ghosh et al., 2012). It was also proved that As content in eggs is directly related with that of drinking water and feed though the biological transmission capacity from body to egg seemed to be low. In a diet study for Hong Kong population, it was observed that mean As content in egg and egg based products is 1.5 µg/kg considering the consumption amount 15 g/person/day (Wong et al., 2013). In case of poultry birds especially broiler chickens, the scenario is somewhat different worldwide. An As containing organic compound named Roxarsone or ROX (3-nitro-4-hydroxyphenylarsonic acid) was used generously in the forage of chickens for more than 60 years. It was used as a growth promoter as well as parasitic disease preventer. It instigated serious As exposure to human health through consumption of chicken meat. In the United States, it has been observed that one person may intake (1.38–5.24) µg of inorganic As from only chicken in a single day assuming mean level of intake i.e. 60 g/person/day (Lasky et al., 2004; Lasky, 2013). Thus, chicken meat was alone responsible to increase lung and bladder cancer risk (Nachman et al., 2013, Liu et al., 2016). As a result, use of Roxarsone was finally banned, the approval of the drug was withdrawn (US FDA, 2014), the manufacturer was suspended. Recently, a study in the poultry farms of Iran has showed that the consumption of eggs from laying hens were safe. Estimated dietary intake of As through eggs from five different strain of hens were considerably low compared to the Provisional tolerable daily intake value (2.14 µg/kg/bw/day) based on Joint FAO/WHO Expert Committee report. The matter of anxiety is, still many countries use roxarsone as a feed additive to animals. A systematic study was conducted in a poultry production of China to measure the effect of As in chicken tissues showing high levels of i-As (0.19−9.7 mg/kg) including ROX (Hu et al., 2017) which is definitely an issue of public health risk. Schimdt (2013) found that organic chicken meat still contains As (geometric mean 0.6 µg/kg) which may be an effect of contaminated drinking water exposure as these chickens are not fed any drug and certified from U.S. Department of Agriculture.


Chapter 2:
Literature review

2.1. Groundwater arsenic contamination: global scenario

Over the years, the presence of As in groundwater is globally labelled as one of the biggest environmental menace to the human population, reported from several countries like Asia, Europe, Africa, North America, South America, Australia etc. (BGS and DPHE, 2001; Nordstrom, 2002; Nriagu et al., 2007). Extensive research in more than 70 countries including Afghanistan, Bangladesh, India, Indonesia, Taiwan, Vietnam, Inner Mongolia-China, Argentina, Mexico, Chile and Thailand showed the variation of groundwater As concentration. Places from Poland, New Zealand, USA, Canada, Bolivia, Hungary, Romania and Japan are also testified with incidence of such poisoning. Around 200 million global people are exposed to potential levels of As (Ravenscroft et al., 2011). It was reported natural As accumulation in soil, surface and groundwater and sea water (Penrose et al., 1977). The contamination is due to leaching from mines or from deltaic alluvial sheds (Smedley et al., 2002). The World Health Organization has reduced the permissible As concentration in drinking water from 50 to 10 µg/L (WHO, 2011a) due to enhanced consciousness of toxicity and lethal consequences of As on human health. Whereas, the highest As concentration found in groundwater of several Asian countries are many folds higher than the allowable limit (**Fig. 4**). It shows that India and Bangladesh are most affected with maximum As concentration 3880 and 4730 µg/L.

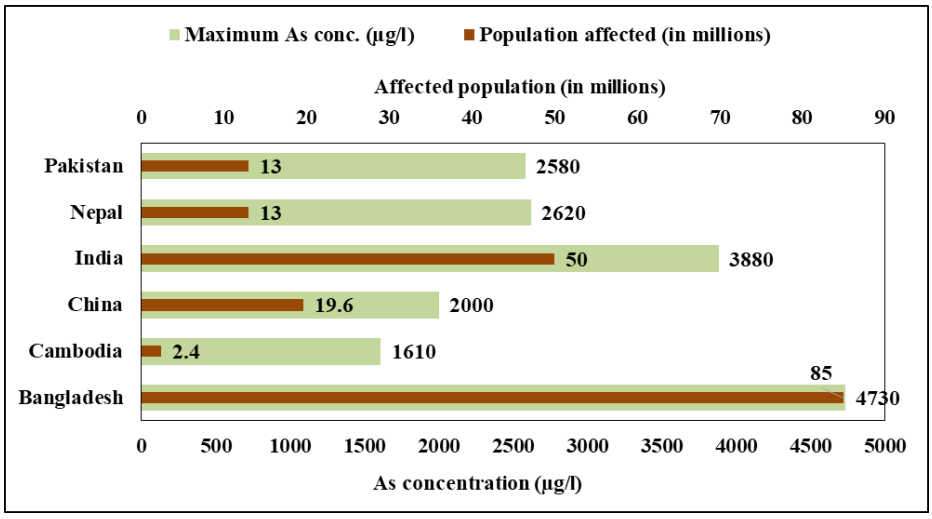


Fig. 4. Maximum arsenic concentration in groundwater of few Asian countries (Shaji et al., 2021**)**

2.2. Groundwater arsenic contamination: Indian scenario

South and Southeast Asian countries are however most severely affected, particularly Bangladesh and adjoining part of West Bengal (Polya and Charlet, 2009; Mukherjee et al., 2008). The GMB river basin (the 13th largest river basin in the world) carries a high sediment bed load. The Bay of Bengal acquires highest quantity of sediments from GMB River that contains quite a few trace elements. An artificial intelligence (AI)-based prediction modelling study by IIT Kharagpur said that nearly 20% of India's total area has toxic As in groundwater (The Hans India, February, 2021). A number of Indian states that fall under the GMB plain (Bihar, Jharkhand, Uttar Pradesh, West Bengal, Assam), Manipur &other North Eastern sites ofIndia, Bangladesh in Padma-Meghna-Brahmaputraplain, and the Terai region of Nepal inIndo-Gangetic alluvial plain are affected with As (Chakraborti et al., 2004). Chandigarh was the first place reported with groundwater As in Indiain the year 1976 in the villages of Haryana, Punjab and Himachal Pradesh (Datta & Kaul, 1976). After few years, groundwater As contamination was reported in the lower Ganga Plain ofWest Bengal was first reported (Garai et al., 1984). Groundwater As contamination in the upper and middle Gangetic plain (Bihar and Uttar Pradesh) was reported by Chakraborti et al. (2003).

2.3. Groundwater arsenic contamination: Incidence in West Bengal

A case regarding subsurface As contamination induced health adversity was first reported in early 1980s in West Bengal by late Dr. K. C. Saha, a distinguished dermatologist from School of Tropical Medicine, Kolkata, West Bengal (Saha, 2000). The group of researchers noticed a number of arsenicosispatients from North and South 24 Parganas districts in West Bengal. Suchobservations initiated the research on finding high As concentration in the groundwater of Bengal delta. The subsequent suffering from As came to limelight during an international conference held in 1995 by School of Environmental Studies (SoES), Jadavpur University Kolkata.Since then, several institutions in Bengal as well as India came forward to perform widespread research focused on As contamination in different geographical strata associated to its health hazards. Work has also been carried out on different remedial measures of As calamity from water, soil and food including awarenessprograms. Early detection of the disease is extremely important as the manifestation on skintakes about 10–12 years to diagnose arsenicosis cases. So, by the time it gets medical attention, As toxicity creates enoughdamage to the body cells. The

incidence of the lethal As in groundwater is described as “the largestmass poisoning in human history” (Smith et al., 2002). In the past 30 to 40 years, the magnitude of As induced sufferings has been amplified by many folds. Currently, nine districts of West Bengal (**Fig. 5**) comprising a population of around 6 million, are severely suffering from As contamination in groundwater (Chowdhury et al., 2000, 2001; Rahaman et al., 2013).

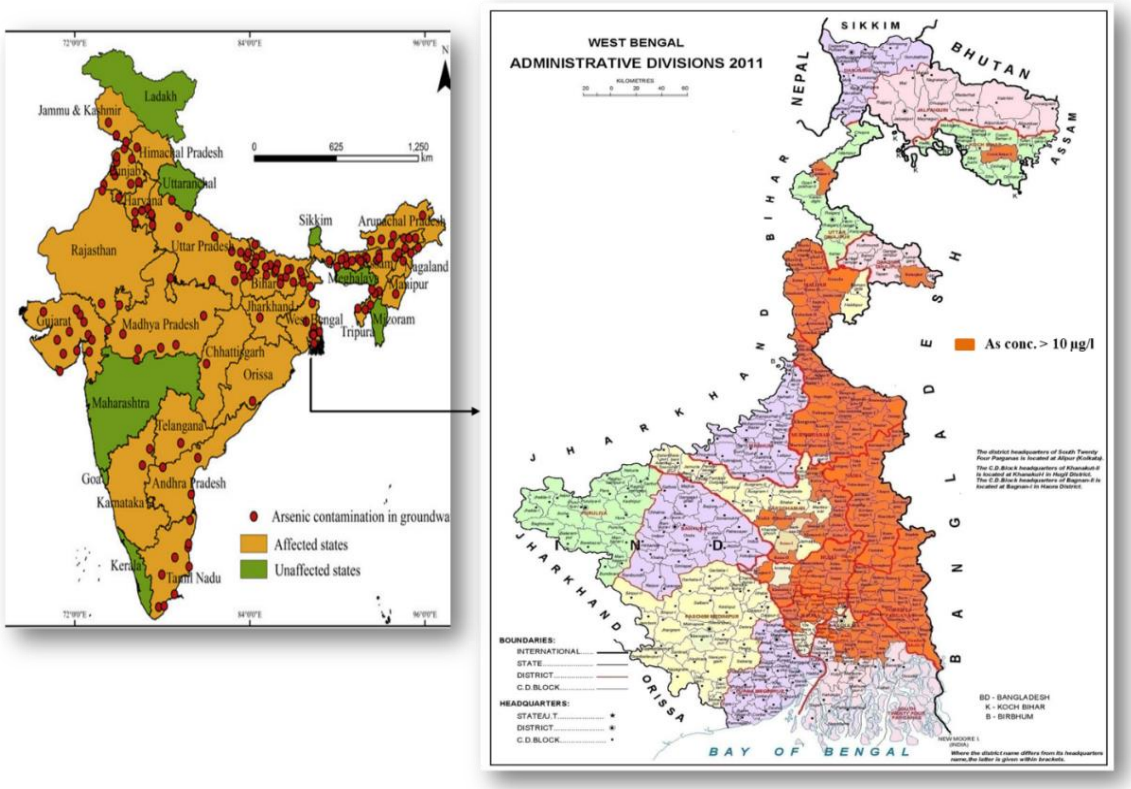


Fig. 5. Groundwater arsenic contamination scenario in India with special reference to West Bengal (Shaji et al., 2021; https://www.appleacademicpress.com/environmental-contaminants-impact-assessment-and-remediation/9781774913963)

2.4. Food chain arsenic contamination

In addition to the contaminated drinking water, food chain As contamination has triggered a serious health concern over the last few years (Samal et al., 2011, Santra et al., 2013). In West Bengal, the largest irrigational production is paddywith approximately 5,900,000 ha cultivation area (Signes et al., 2008). The Bengal populationmajorly depends on rice as a basic diet and interestingly rice grain accumulates more As than other frequently harvested crops (Mondal and Polya, 2008, Kumarathilaka et al., 2019, Williams et al., 2007). Accumulation of As in paddy grains fundamentally differs with season, variety, As concentration in water and soil which is elaborately described in two recent reports along

with As accumulation and distribution in the growth phases of paddy plants (Chowdhury et al., 2018a, 2020a). Paddy cultivation requires huge amount of water, which is fulfilled by As contaminated groundwater during summer (Chowdhury et al., 2018a) and also in the monsoonwhen rainwater is insufficient (Chowdhury et al., 2020a). Furthermore, the post-harvesting mechanism of raw rice grain has caused an additional Asburden in these areas where the parboiled rice and rice-by products are detected with higher As concentration (Chowdhury et al., 2018b, 2019). While comparing the As species in rice varieties, it is well established that inorganic As dominates the methylated As (DMA, MMA etc.) in a high ratio (Islam et al., 2017b, Pal et al., 2009, Roychowdhury, 2008). It is appropriate to cite that inorganic As is way more toxic than any of its methylated species (Signes Pastor et al., 2008, Jomova et al., 2011). This is not an only concern for the local inhabitants because the crops cultivated here, gets transported in the widespread urban areas (Biswas et al., 2019). A household as well as market basket survey of food materials from different locations of apparently unexposed Kolkata city shows that the urban population is also under the same threat of As toxicity though at a lower degree. Even, the cooked rice is another point of health concern in Bengal delta where As concentration varies with cooking method, cooking water, rice and water proportion (Chowdhury et al., 2020b; Mondal et al., 2010a; Mandal et al., 2019; Rahman et al., 2011; Sengupta et al., 2006). The distribution of As accumulation in different parts of rice while cooking is brilliantly depicted by Chowdhury et al. (2020b); in uncooked and cooked rice, cooking water, and gruel. Major part of the total As content in cooked rice is also contributed by i-As species (Chowdhury et al., 2020b, Halder et al., 2013, 2014, Roychowdhury, 2008, Laparra et al., 2005). The suggested intake value of inorganic or total As is 3.0 μg/kg bw/day, which is revised after the 72nd meeting by the Joint FAO/WHO Expert Committee advised PTDI value (2.1 μg/kg bw/day) was withdrawn (EFSA, 2014, JECFA, 2011, WHO, 2011). In most of the As exposed populations, researchers have observed that the average daily dietary intake rate is much higher than the recommended limit (Bhowmick et al., 2018, Halder et al., 2013, Roychowdhury, 2008, 2010). Therefore, As contamination in drinking water and foodstuffs, being a potential hazard to the mankind, needs to be immediately taken care of (Das et al., 2009, Mazumder, 2003, Rahaman et al., 2013).

2.5. Adverse effects of arsenic on human health

Being a group I carcinogen (IARC, 2012), As exposure for a prolonged period causes cancer in different organs which in turn increases mortality (Abdul et al., 2015; Ng et al.,

2003). In highly endemic areas, every year numerous patients who have been identified as having been exposed to As have had skin carcinoma (Chowdhury et al., 2000; Mazumder et al., 1998). In different districts, As toxicity and the sufferings of people vary depending on the water As concentration range. Analysis of the biological samples of the inhabitants shows the extent of As toxicity in their health (Das et al., 1995, Rahman et al., 2001, Roychowdhury, 2010). Continuous intake of As for a short period of time may cause abdominal pain, diarrhea, or vomiting, while the most familiar manifestations of chronic As exposure are lung dysfunction, persistent cough, obstetric problems, thickening of the skin, hyperpigmentation, keratosis, and melanosis (Rahman et al., 2009; Ratnaike, 2003). It causes hyper-pigmentation, stiffening of the skin, keratosis, melanosis, squamous-cell epitheliomata, black-foot disease, and gangrene (Abdul et al., 2015; Chowdhury et al., 2017; Rahman et al., 2009, Ratnaike, 2003). The inhabitants residing in Murshidabad have been suffering from chronic As toxicity since many years which includes severe arsenical skin manifestations (Chakraborti et al., 2009; Rahman et al., 2005b, c). Arsenic poisoning imposes different health hazards including skin problems, respiratory troubles, cardiovascular diseases, nerval disorders, renal issues etc. (Mazumder, 2008). Arsenic toxicity is again known to disturb the reproductive system badly. Miscarriage, pre-mature birth, perinatal and prenatal mortality are few examples (Chakraborti et al., 2013). In the early 1980s, As toxicity was reported first in India from West Bengal (Mazumder et al., 1998). It is not only adults who are affected; reports say that, worldwide, children, infants, and young adults are also facing a cancerous risk from As contaminated drinking water (Baig et al., 2016; Brahman et al., 2016; Singh and Ghosh, 2012). Even a low level of As exposure can prove to be a health risk in children with many disorders (Biswas et al., 2018). Lately, children of endemic areas of West Bengal (Gaighata, North 24 Paraganas district) have been sub-clinically affected and have a considerable future cancer risk (Joardar et al., 2021b). Earlier, children aged <9 years from South 24 Paraganas, West Bengal, India were recountedwith skin pigmentation and keratosis after As exposure, although to a lower degree compared to adults (Mazumder et al., 1998). Children from Bangladesh of age between 4 to 15 years were also reported with dermatological symptoms due to consumption of As-contaminated drinking water (Rahman et al., 2001; Watanabe et al., 2007). Acute or chronic As exposure on children throughout the world include slow growth rate, weight loss, impaired intelligence, and loss of memory (Calderon et al., 2001; Wasserman et al., 2018; Wright et al., 2006). As exposure may also have affected the cognitive function of children aged 68 years in Mexico (Rosado et al., 2007). Children

with prenatal and early-life As exposure (above 500 μg/L in drinking water) were detected to have pulmonary difficulties in Bangladesh (Smith et al., 2013). In-utero As exposure also caused a reduction in the thymic development of children with increasing morbidity in Bangladesh (Raqib et al., 2009). Higher mortality due to different forms of malignancy was also reported in early adults with in-utero and childhood As exposure in Chile (Smith et al., 2012). Furthermore, in developing and under-developed countries, malnutrition of children influences As toxicity and lessens their quality of health (Calderon et al., 2001; Milton et al., 2004; Rahman et al., 2001). Biswas et al. (2019) and Joardar et al. (2021a, b, 2022) showed that As exposure is constrained neither to As prevalent areas nor to any exact age. The viciousness of As exposure hinges onseveral factors such as dietary intake over food and drinking water (Joardar et al., 2021a; Kumar et al., 2016; Roychowdhury et al., 2003), genetic susceptibility (Paul et al., 2015), age and sex (Rahman et al., 2006), exposure occurrence, nutritional status, socio-economic status (Brima et al., 2006; Chakraborti et al., 2018; Rahman et al., 2018; Hata et al., 2007), etc. School going children between 5 and 15 years seemed to be affected with As toxicity in both As exposed and apparently unexposed areas of Bengal, reported by Joardar et al. (2021b). Joardar et al. (2021c) investigated health risk evaluation caused by As among few mothers (aged between 23 and 31 years aged) and their children (from 7 months to 4 years) in an As-exposed village of North 24 Parganas district, and showed that the populations are sub-clinically affected. The adults in the differently As exposed villages own a higher urinary As concentrations or hair As concentrations compared to children (Joardar et al., 2021a). But Bibi et al. (2015) proposed that the children and aging people have higher Asaccretion in hair samples compared to adults, that may be a result of feebler metabolism and higher proneness for ailments. Although, both age and gender were reported to be insignificant variables inassessment of hair As concentration as told by Das et al. (2018); Hadi and Praveen (2004) and Wu and Chen (2010). According to Bozack et al. (2018) and Torres-Sánchez et al. (2016), As metabolism in human body systems are ruled by sex and age differences.

2.6. Exposure of arsenic in livestock

The literature study is imperative to assess the geo-chemical As exposure in heifers from As endemic area through their biomarkers, particularly domestic cattle and goats compared to the control area livestock. Cattle, goats, fowl and hens which raise in the households of the As endemic sites are anticipated to be exposed with As toxicity through daily diets.

The fodder of these livestock includes whole crops and crop remnants e.g. rice straw, rice husk, crushed rice grain, maize, corn dust and husk, mustard cake, wheat chaff and pea chaff that reported with considerable As concentration (Rahaman et al., 2013; Rahman et al., 2008). Livestock typically do not reveal any noteworthy As mediated skin manifestation, but agonize having sub-clinical toxicity (Mandal, 2017). Cattle were more commonly found with As befouling amid all the other animals both acutely and chronically (Selby et al., 1977). Miranda et al. (2005) reported that nonetheless, the toxicity level was low in calves, liver and kidney were found with the highest exposure. After metabolism, around 80% of i-As and other organic As species were assimilated in liver and kidneyof the mammals and then eliminated by urine and faeces (Kicin´ska et al., 2019). Although apparently most of the livestock excrete As rapidly informed by People (1964). Prolonged consumption of As persuades a heavy deposition in theectodermic tissues likehair and nail (Eisler, 1994). Arsenic passes through few metabolictransformationsin mammalian animals like arsenate to arsenite followed by the methylated species, monomethylarsonicacid (MMA) and dimethylarsinic acid (DMA). The reduction to arsenite from arsenate occurs by glutathione (GSH) and the thiol groups, while the enzyme methyltransferase performs methylation (Roy et al., 2013; Vahter, 2002). Nevertheless, for all the mammalian species, methylation capacity of i-As isnot same (Abernathy et al., 1999; Vahter, 1994). Sattar et al. (2016) and Ventura-lima et al. (2011) explain the metabolism of As in mammals systematically. Arsenic readily traverses the placenta barrier in mammals, resulting in exposure in both fetus and the mother (Mandal, 2017). Considerable amounts ofAs in the animal sourced products and parallel human health threatthrough ingestion of these comestible products have been assessed by Ahmed et al. (2016), Joseph et al. (2015) and Islam et al. (2014). Cow milk is essentialfor children foodas a potential source of protein, calcium, iodine, vit-A and other micro-nutrients; reported in Benelam et al. (2015) and Ferna´ndez et al. (2014). Abedin et al. (2002) revealed that As-contaminated milk is secreted by the lactating cows who consume As-contaminated food for an extended period. Similarly, eggs produced by hens grown in As prone areas were found with substantial As in Bangladesh (19.2 µg/kg) and Mexico (26 µg/kg) (Ghosh et al., 2012; Del Razo et al., 2002). Devi et al. (2014) reported that animal meat isalso enriched with proteins, minerals, vitamins, micronutrients and fats speciallyconjugated linoleic acid and omega-3 fatty acid. Adequate As in chicken, duck, goats or cattle flesh was also observed by Islam et al. (2014) and Shaheen et al. (2015).

Recommendation of i-As in total dietary intake for human is 3.0 µg/kg bw/day in accordance to WHO (2011b).

2.7. Mitigation options of arsenic contamination

Arsenic contamination in drinking water, soil and food needs to be remediated for the safety and good health of human beings. Adsorption is a suitable way to reduce the extent of As contamination in drinking water. Application of activated carbon is expensive and carbon can eliminate few milligramsof metallic ions per gram of activated carbon and this process deals with a huge sludge disposal problem. Researches are carried out to produce a low-cost adsorbent. Activated alumina is also an effective adsorbent in situ to extend the useful life. There are few easily available and affordable adsorbents like clay, silica, sand, etc. which can be redeveloped in situ. Regrettably, theseadsorbents have lesser efficiency than the otheradsorbents. Other water contaminants can also disable the sorption efficiency of such adsorbents. Organicpolymers cam also be used asdecent adsorbents with in situ regeneration ability. Various researches are carried out using bio-sorbents i.e. natural adsorbents like fruit peels and agricultural wastes (Yeo et al., 2021). Apart from adsorption, there are various other conventional processes whose efficacy, practical applicability and procedural achievability are being researched since past few years. Besides, drinking water mitigation measures are also needed from soil. For production of benignrice grains with less As concentration, application of extenuation processes is necessary in As contaminated rice agro-ecosystems. Potential agronomical, physico-chemical and biological practices are applied to lessen both the bioavailability of As in the paddy soil and significant uptake and assimilation in rice grains. **Fig. 6** depicts a short summary of different mitigation processes from contaminated drinking water, soil and rice.

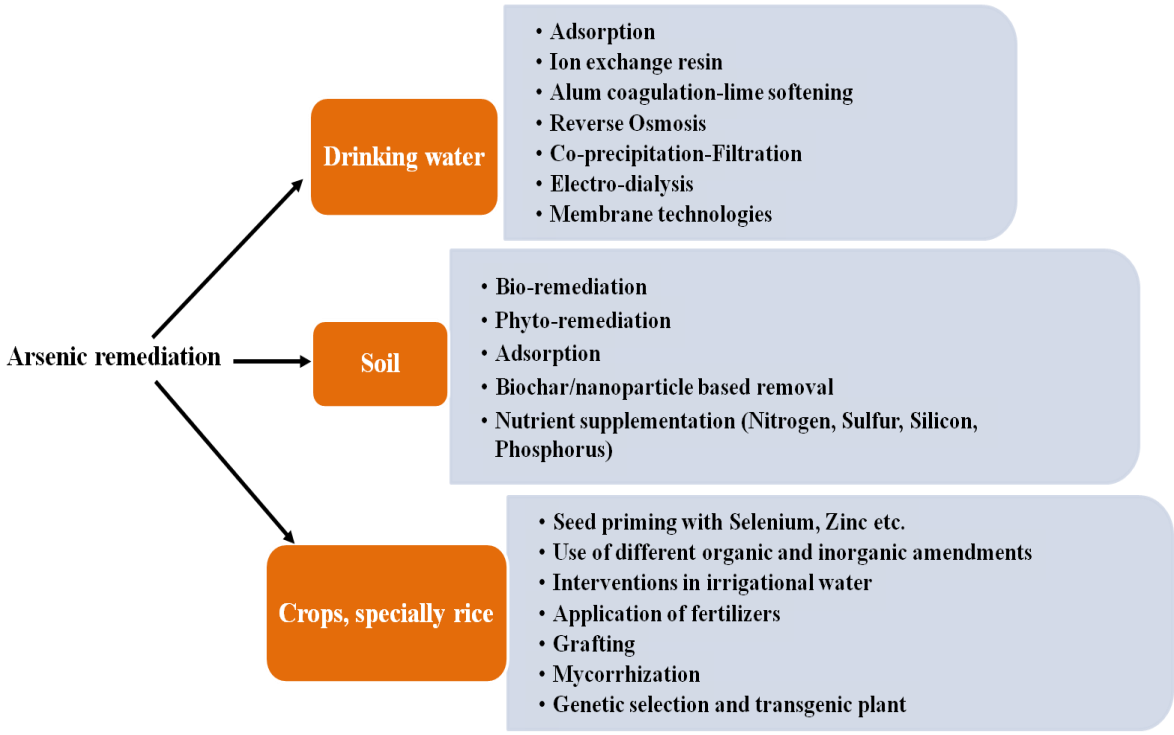
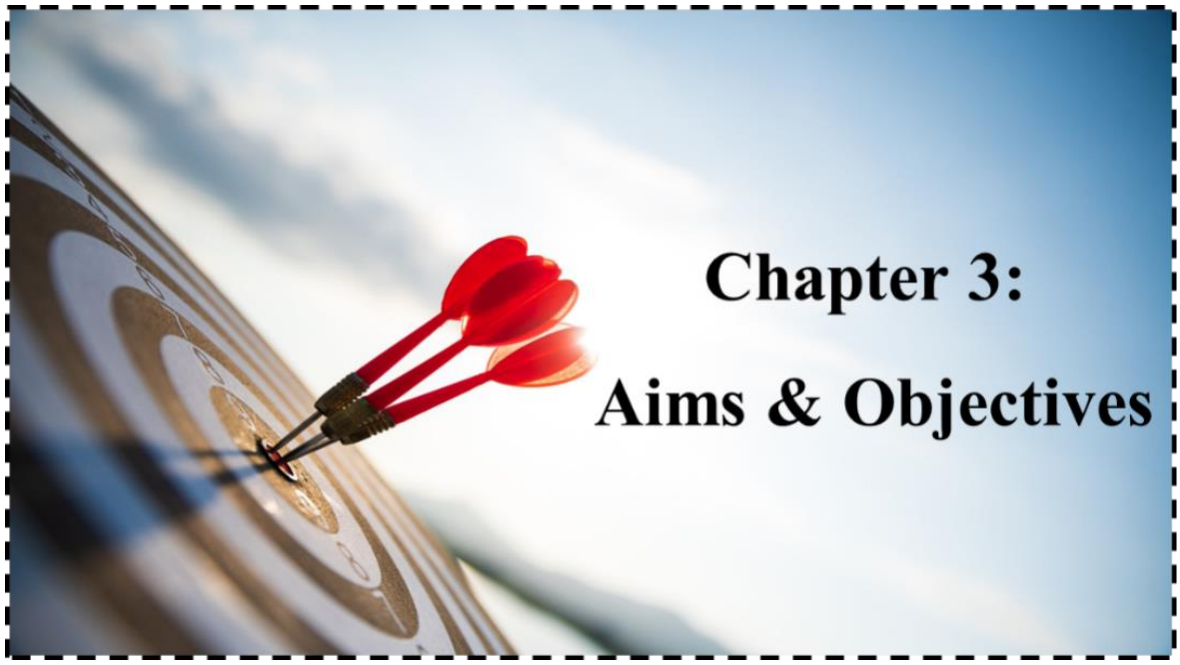


Fig. 6. Conventional methods of arsenic remediation from different spheres

Several researches elaborately discussed the pros and cons of As remediation from different strata of environment like water, soil and food grain (Alka et al., 2021; Min et al., 2008; Mohan et al., 2007; Mohamed et al., 2012; Mridha et al., 2021, 2022a, b, c; Ray et al., 2021).


Chapter 3:
Aims & Objectives

Arsenic exposure has crossed nearly 40 years after its first incidence in West Bengal. During these years, several mitigation measures have been undertaken to reduce As contamination. However, the problem has not been eradicated, even, in few cases it is magnified. Therefore, the present research study aims to:

- Investigate the current groundwater quality of selected As exposed sites for drinking, domestic and irrigational purposes after so many years of its exposure.
- Evaluate the quality of all kind of available alternate drinking water sources, assess the performance of the As removal plants and finally suggest the safest source of drinking water.
- Illustrate the mode of interaction of As with other essential and non-essential elements in groundwater and to evaluate is effect in daily diet.
- Examine the role of As in rice grain from groundwater and soil through comparative approach between As content and concentration.
- Evaluate the extent of Asexposure in human population based on different hypotheses (age, sex and dietary intake) as well as identify the most significant factor for their futuristic health risk.
- Estimate the effect of Asexposure on domestic livestock followed by human through consumable animal by-products and consequent environmental risk.
- Promote an inexpensive, easily accessible solar oxidation de-arsenification method at domestic level using iron and citric acid enriched organic components.


Chapter 4:
Materials & methods

4.1. Study Area

West Bengal, India, is widely affected by groundwater As pollution for past several years. Nine districts in West Bengal i.e. Malda, North 24 Parganas, Nadia, Murshidabad, South 24 Parganas, Kolkata, Purba Bardhaman, Hooghly, and Howrah have high As pollution in groundwater currently. Arsenic contamination in groundwater in the eastern part of Bhagirathi River is a burning public health issue so far and well documented by a lot of researchers. The study area of my research work comprises evaluation of groundwater quality and investigation of health exposure in several sites of West Bengal, India including Nadia, Bardhaman, Murshidabad, North 24 Paraganas. According to the acquaintance of samples, the research work was carried out further on selected control sites of West Bengal (Budge Budge in South 24 Parganas, Belur in Howrah and Madhyabar inPingla block, West Midnapore). Madhyabar, an As unexposed agriculture based village, whereas Budge Budge and Belur are As unexposed urban and industrial towns.

4.1.1. Nadia

All the 17 blocks of 'Nadia' district is covered under the research work to evaluate the quality of groundwater for drinking or domestic purposes. The approach on health exposure from As has been carried out with special reference to two worst As exposed blocks named Chakdah and Haringhata (**Fig. 7a**). Nadia district, approximately 46 ft. above the sea level (lat: 22°47′10″ N and long: 88°55′65″ E) lies on the eastern bank of the Bhagirathi River and covers about 3927 km^2 area. Nadia is bounded by Bangladesh, Bardhaman, Murshidabad and North 24 Parganas districts in the East, West, North and North West, South and South East respectively. This district consists of 2 notified areas, 8 municipalities, and 17 blocks including 187 Gram Panchayats that consists of nearly 1307 villages. Rahman et al. (2014) informed that the population of Nadia is approximately 51.7 lakhs thatvastly depends on tube-well water and only some piped lines water supply. Nadia grows seasonal crops throughout the year for its sufficient groundwater and new alluvium soil. The economy of the district depends mainly on agriculture as all the important crops

are commercially grown in this district. Rice, jute, oil seeds, wheat and pulses are the majorly grown crops throughout the year and the cropping intensity was found to be 249 %. The total productivity of rice, wheat and potato are reported as 2665, 2217 and 22860 kg/ha annually (average of 2004, 2005, 2006, 2007 and 2008). The source of irrigation in this district mainly covers open wells (n =33), bore wells (high discharge tube well, n= 680), lift irrigation schemes, shallow tube-wells etc. Net irrigated are and rain-fed area are 209.6 and 479.8 thousand ha respectively. Approximately 88 % irrigational area is reported as 'critical' according to the groundwater utilization (Agriculture Contingency Plan for District: NADIA, 2011). Groundwater of Nadia is highly polluted with As, as informed in various researches (Das et al., 1996; Mazumder et al., 2010; Rahman et al., 2014; Roychowdhury et al., 2008). According to Rahman et al. (2014), groundwaterin all the 17 blocks of Nadia is As contaminated where approximately 51.4% and 17.3% of the tube-wells had As above 10and 50 μg/L (Rahman et al., 2014). Haringhata (22.9632°N, 88.5673°E), a municipal town under Haringhata block, Nadia has a total population of 3,989 (Census, 2011, https://www.census2011.co.in/data/town/322410-haringhata-farm-west-bengal.html). Chakdaha is another town and a municipality in the Kalyani subdivision of the Nadia district (23.08°N 88.52°E). As per the census of 2011, the Chakdaha Municipality has a populace of 95,203 where 48,047 are males and 47,156 are females. Moreover, health effects of two different age groups of school children (9-10 years and 14-15 years) studying in 'Laupala Kalpataru Primary School', 'JaguliJunor Basic School' and 'Laupala Kalpataru High School' from Chakdah and Haringhata Municipality area under the jurisdiction of Nadia district have been extensively studied under my research work (**Fig. 7b**).

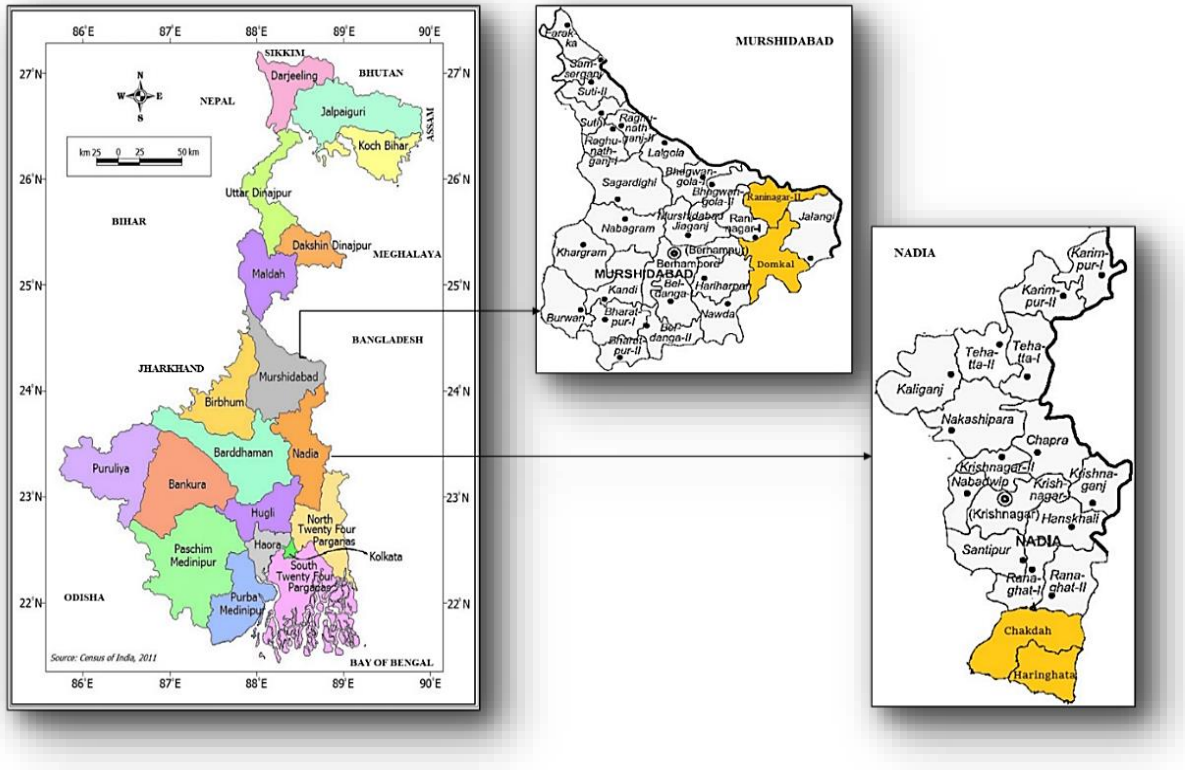


Fig. 7a. Block distribution in Nadia and Murshidabad district

Fig. 7b. Selected schools of Nadia for health effect study on children

4.1.2. Bardhaman

Secondly, Bardhaman is another district in West Bengal covering 7024 km^2 area (latitude 23°13′57.0468″ N and longitude 87° 51′48.3084″ E). The district lies between the rivers

Ajay and Bhagirathi-Hooghly. The geographical boundary of this district involves Birbhum and Murshidabad on North, Nadia on east, Hooghly on south, Jharkhand and Purulia on West. The district has 4 sub-divisions named Asansol, Durgapur, Burdwan-Sadar (North-South), Katwa and Kalna. The district consists of 32 community development blocks, 9 municipalities, and 2 municipal corporations which encompasses approximately 6.91 million population (Census, 2011). All the 32 blocks of Bardhaman district are covered under the research work to evaluate the quality of groundwater for drinking or domestic purposes (**Fig. 7c**). It is the only district in West Bengal that has both in industry and agriculture; about 58% of the total population belongs to the agricultural population and the non-agricultural division accounts for the remaining 42 %.With a cropping intensity of 184% the total productivity of major crops like rice, wheat, jute and potato are 3012, 2313, 3019, 21674 kg/ha.The source of irrigation in this district mainly covers canals and lift irrigation schemes (n= 1116) etc. and total irrigated area is 331.6 thousand ha. 5 blocks are reported as 'semi-critical' according to the groundwater utilization (Agriculture Contingency Plan for District: Bardhaman, 2011). Arsenic contamination is reported in 5 blocks especially Purbasthali I and II (Biswas, 2010; Ghosh and Mukherjee, 2002; Nag et al., 1996).

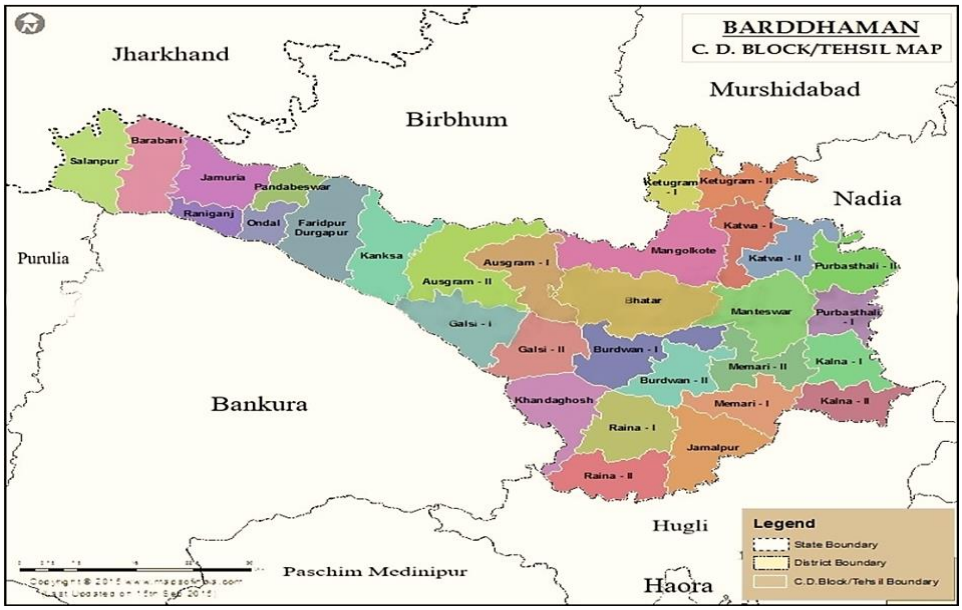


Fig. 7c. Block distribution in Bardhaman district

4.1.3. Murshidabad

Murshidabad district is situated at the eastern bank of the stream Bhagirathi; the border between West Bengal and Bangladesh. The hydrogeology of Murshidabad delta has a thick

granular region (150–250 m) with a small clay layer comprising unconfined aquifer with high As concentration (Chakraborti et al., 2009). Murshidabad is an agriculture dependent district with 204.3 thousand ha of net irrigated area and 408.6 thousand ha of rain-fed area where the cropping intensity is reported as 245%. The source of irrigation in this district mainly covers canals, tanks (n= 11024), lift irrigation schemes (n= 917) and other sources. Rice, wheat, jute, potato are the most cultivated crops and vegetables with a total annual productivity rate of 2678, 2295, 3048 and 15225 kg/ha. The major source of irrigation in this district includes canals, tanks, lift irrigation schemes, shallow tube-wells etc. (Agriculture Contingency Plan for District: Murshidabad, 2011). Out of the 26 community development blocks (an administrative unit in district for rural development) of the district, the research study covered Raninagar-II and Domkal blocks which falls under the jurisdiction of Domkal sub-division (Fig. 7a). Raninagar-II is located at 22 m above the sea level and is circumscribed by Rajshahi district of Bangladesh. The block covers an area of 206 km^2 including 9-gram panchayats, 36 mouzas, 127-gramsansads or village councils, and 30 colonized villages covering a population of approximately 1.91lakhs (Census, 2011). Domkal block owns 305.19 km^2 area and 1 panchayat samity, 13-gram panchayats, 249-gramsansads, 87 mouzas and 77 colonized villages. Domkal and Raninagar-II block had a populace of 363976 and 190,885, all being rural (Census, 2011).

4.1.4. North 24 Parganas

North 24 Parganas extends in the tropical zone of West Bengal; surrounded by Nadia at north, Bangladesh at north and east, South 24 Parganas and Kolkata at south and Howrah, Hooghly atthe west. The district is the most populous one in West Bengal and a part in the Gangetic domain, at the east of the Hooghly River and the groundwater is extensively contaminated with As (Chakraborti et al., 2001). Groundwater is mostly As affected in several blocks like Gaighata, Deganga (Chowdhury et al., 2018; Joardar et al., 2021a). Out of the total 22 blocks in North 24 Parganas, Gaighata (22.934521°N, 88.730754°E) is designated as one of the most badly As affectedone (Chakraborti et al., 2009). Gaighata has a total population of 330,287, whereapproximately 80.4 % population is rural (Census, 2011). Roychowdhury (2010) conveyed that 107 mouzas covering 13-gram panchayats in Gaighata block have As-polluted groundwater with more than 10 µg/L and an average As concentration of 113 µg/L. The research study covered work on As exposed few blocks of this district: Gaighata, Bongaon, Baduria etc. (**Fig. 7d**). Different villages across Gaighata, have also been chosen assites of exposure on domestic livestock. Also, a number of 25

arsenic-iron removal plants located in different gram panchayats of Gaighata, have been studied under this research workto evaluate the efficiency of applied mitigation strategies (Fig. 7c).

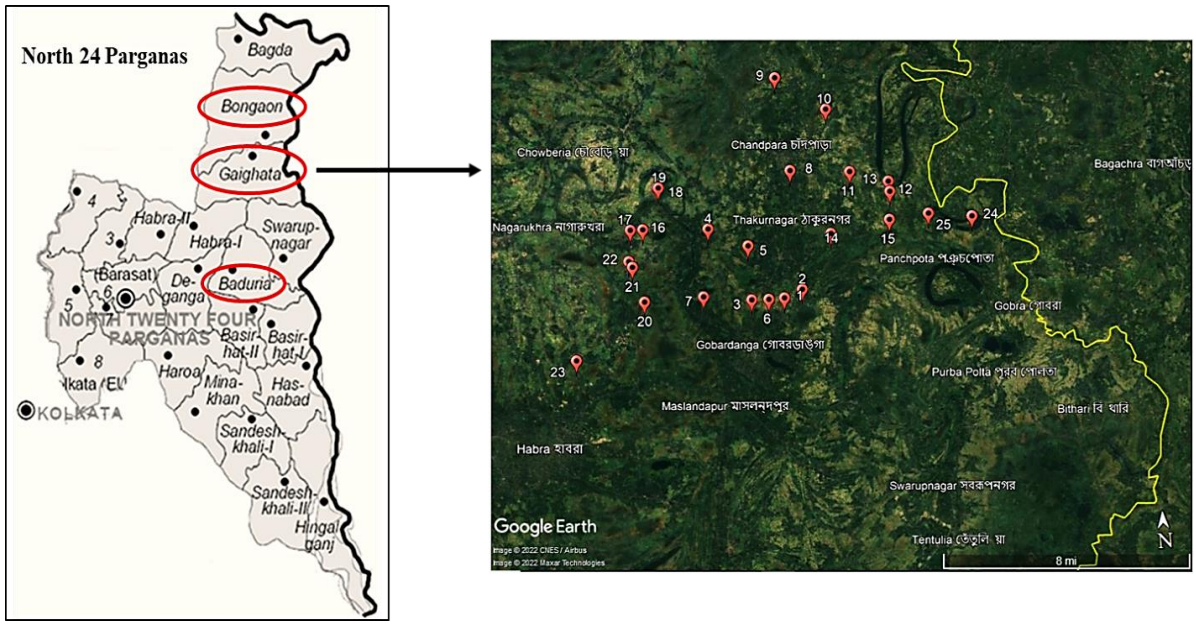


Fig.7d. Selected study sites of North 24 Parganas

4.2. Collection of sample and preservation

4.2.1. Sample type: Water

The research study has been carried out primarily by collecting groundwater samples used for drinking and cooking purposes in the households of the studied areas. It followed by collection of all kind of available drinking water samples in the study sites as well as irrigational water samples in the agricultural fields where cultivation takes place regularly. Collection of samples were made induplicates in pre-washedair-tight polyethylene containers (50 ml and 250 ml) from the different water sources e.g. deep tube-well, shallow tube-well, pipeline supply, dug wells, treatment plant waters etc. The GPS or 'Global Positioning System' information has been considered to identify the sample source accurately with exact latitude and longitudes for further process, if needed. The facts about depth of the tube-wells have been noted from the household level of local inhabitants, local panchayat (governmental) level and farmers working in the agricultural fields. In the containers of 50 ml, water samples were stored adding 0.1% (v/v) of concentrated HNO_3 as preservative to estimate As, Fe, U and the micro-nutrients. In the other 250 ml container, same samples were collected without adding any preservatives to determine other physico-

chemical parameters. Water sample containers were transported to the laboratory by keeping the samples in an ice box as early as possible andkept at 4°C till estimation.

4.2.2. Sample type: rice grain

Different varieties of whole paddy grain and rice grain were collected from the farmers' houses in the study areas. Also, for the direct field exposure and for the study of As translocation in plant pedicels, paddy grains were collected directly from selected fields with the help of the farmers. Whole paddy grain samples were divided manually into three different parts in accordance to height: top, mid and bottom. For, food chain As exposure study, cooked rice sample and gruel (spare water from cooked rice) samples were collected from the families while rice was being prepared. During the time of sample collection, information about the samples has been gathered. The rice samples were carried to the laboratory storing in separate zip-lock packets. Each water sample used in rice cooking process were collected in sterilized polyethylene ampoules and preserved by 0.1% (v/v) concentrated HNO_3 (69%) and kept at 4 °C till estimation. The gruel samples were collected too individually in fresh containers without adding any preservatives and stored for analysis.

4.2.3. Sample type: Biological (Human)

Urine, scalp hair and nail samples had been collected from the respected family members for As exposure study in human. Information regarding their age, sex and duration of exposure, food habit, intake rate etc. were carefully noted. Nail clippings from tip of the fingers and hair samples from scalp were collected directly with use of ceramic blade and stainless-steel scissors respectively. At room temperature, these biological samples were conserved distinctly in sealed zip-lock packets. Washing of the hair and nail samples were done systematically using double distilled water, and then with acetone by a magnetic stirrer to remove the outer adsorption of As on the samples. The washed samples were kept in glass beakers separately and dried at 50 °C overnight in a hot air oven. Collection of the urine samples weredone at spot in 20 ml polyethylene bottles without any preservatives added. The urine samples were stowed at – 20 °C before analysis. The samples were diluted by 1:1 with double distilled water and filtered before analysis to remove colloidal particles and lessen matrix effects.

4.2.4. Sample type: fodder, excreta, soil (livestock)

Protocols for sample collection, preservation and preparation for livestock exposure work are shown in details in **Fig. 8**. For As exposure study in domestic livestock, cattle, goats and hens which are reared in the villagers' houses have been accounted for the currentwork. Groundwater samples that for drinking, foodstuff samples like mustard cake, crushed grains and husks, biological sample like urine, faeces and tail hair, edible animal product sample like milk, egg, chicken, goat liver and flesh were collectedseparatelyfrom the study areas and stored carefully. At the time of milking, cow milk samples were collected separatelyin 100 ml plastic containers and similarly from the cans of milkmen, kept in ice-cold condition during carrying to the laboratory and finally stowed at - 20 °C till analysis. Whole cow milk-arsenic fractionation study had been performed on few of the selected milk samples. Collection of egg samples were done from different hens of the families, while liver and meat samples (chicken and goat) were collected from the local butcher's shops of the rural community. Also, Surface soil samples were collected from different agricultural fields, fallow lands and cowsheds of the As exposed sites of West Bengal to estimate the extent of As contamination caused by the excreta of livestock. This will be helpful to evaluate environmental risk as well.

The solid samples were collected in separate polyethylene zip-lock systems. Milk and milk-based products such as curdled milk, sweets and syrup of wet sweets were collected from three sweet-shopsof the As exposed villages. The egg samples were cooked with As -free tap water and sharedcarefully into two parts; albumen and yolk; and kept in a fridge at wet condition. Sample collection and preservation process of urine and tail hair are same like humans and has been discussed in the earlier section. The cow dung and soil samples were placed separately in watch glasses and dehydrated in a hot air oven for 48 hrs at 45 - 55 °C to remove the moisture content. The dried samples were stored in separate polyethylene zip-lock bags. Next day, they were grinded in a mortar and pestle and sieved carefully and stored again. For milk fractionation work, the raw milk samples were poured in a 50 ml sterilized centrifuge tube, homogenized well through a vortex (REMI CM 101) and centrifuged at 4000 rpm using (REMI R-8C) for 15-20 minutes. The homogenized solution then got separated into two parts, fat and skimmed milk. The upper fat portion was taken apart and skimmed milk portion was again ultra-centrifuged at 15000 rpm for 1 h; a substantial amount of skimmed milk was also taken apart for total As estimation. Finally, the whey portion (supernatant) and casein (pellet) were separated carefully. The

milk fat and casein sample were collected and placed in individual watch glass and dehydrated in the hot-air oven at 50 to 55 ºC; the whey portion was kept in a salt-ice mixture after immediate collection, stored at -20 ºC. The next morning, defrosting was done of the whey, skimmed milk and whole milk samples at room temperature and those were homogenized customarily by the shaker for 10 minutes before digestion.

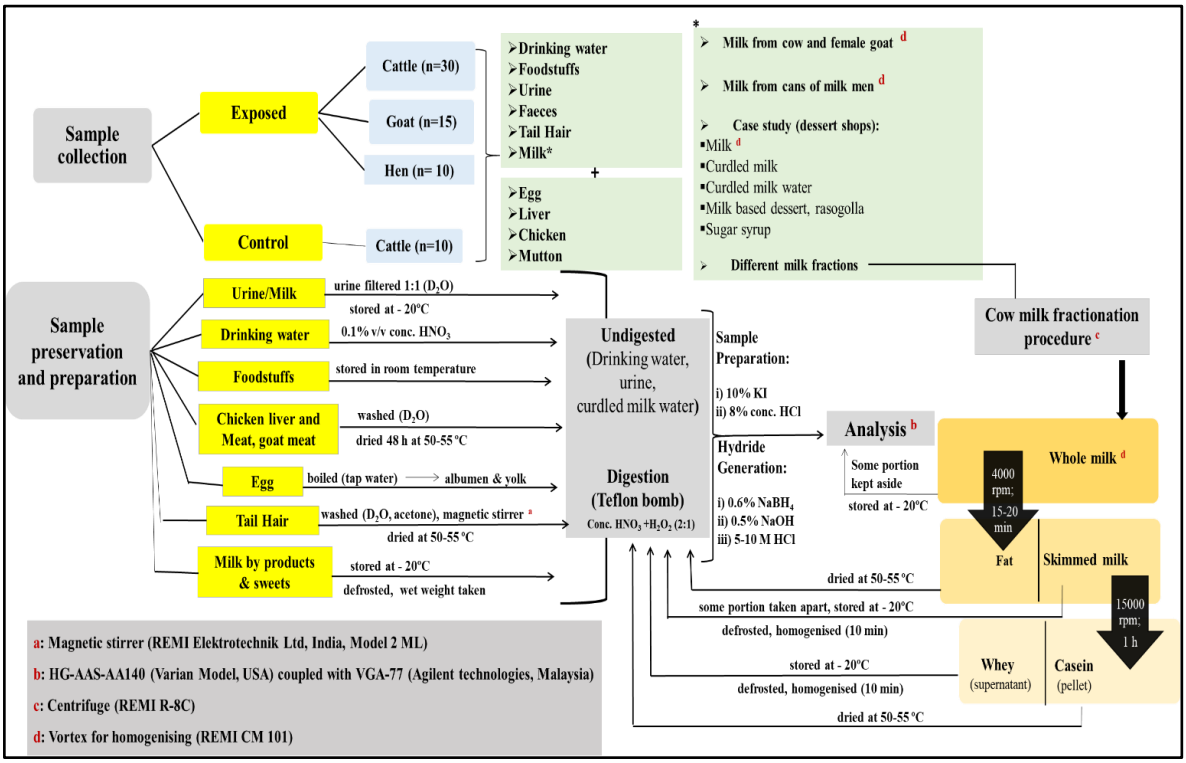


Fig. 8. Protocols for sample collection, preservation and preparation

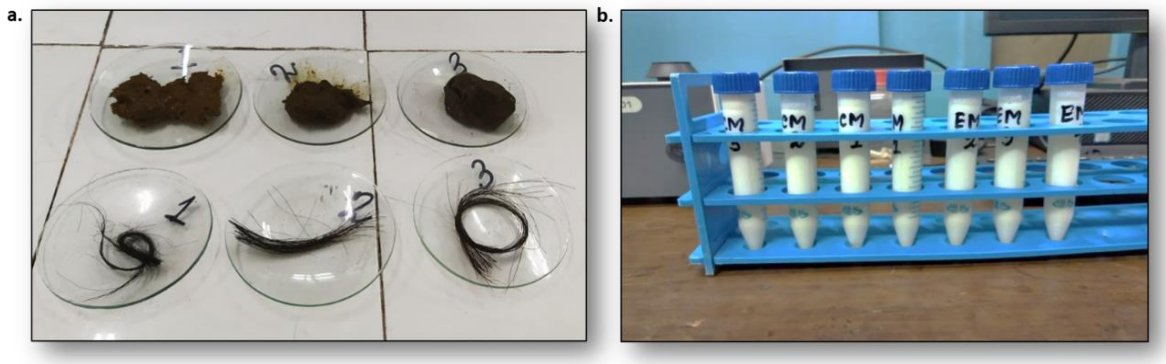


Fig. 9. Livestock sample: (a) Cow dung and tail hair, (b) cow milk

4.3. Sample processing

4.3.1. Digestion

No digestion process was followed before estimating As in water and urine samples only. About 0.02-0.2 g of all kind of individual solid and semi-solid samples (dry weight) were placed in Teflon containers; 69% concentrated HNO_3 and 30% (v/v) H_2O_2 were added in a proportion of 2:1. The teflon containers were kept into their particular bombs and placed in an oven at ~120 °C for 6 hr. Subsequent to cooling of the bombs, the digested samples were placed on hot plate at ~90 °C for 1 h for evaporation to a final volume of 2 - 5 ml with double distilled water and filtered through a millipore suction filter (0.45 mm). Then the filtered samples were stowed at 4 °C till analysis was done. 50 % of the samples were digested only in a hot plate method.

4.3.2. Process of extraction of As species and instrumentation

Arsenic species were extracted by addition of a 1:1, (v/v) mixture solution of methanol and water. The detailed process of the analytical work for the As species (MMA, DMA, inorganic arsenic) extraction has been stated previously in Roychowdhury (2008) and Signes-Pastor et al. (2016). Speciation was done from the 'Global Centre for Environmental Remediation (GCER)', The University of Newcastle, Australia. Instrumentation details for As speciation using high performance liquid chromatography (HPLC) coupled with inductively coupled plasma-mass spectrometry (ICP-MS) has been described earlier (Islam et al., 2017; Roychowdhury, 2008).

4.3.3. Estimation of essential and non-essential trace elements and other heavy metals

Essential and non-essential micro elements including heavy metals the drinking water samples were analyzed using a fast and productive Atomic Absorption system, attached with premium optics, UltrAA lamp looms (standard IS method), (Agilent 280 FS AA model) at National Test House (NTH), Dept. of Consumer Affairs, Govt. of India, Salt Lake, Kolkata as well as ICP-OES through peri-pump sample introduction method (model: G8422A, serial number SG20400806, auto-sampler type SPS4), Agilent India, New Delhi.

4.4. Laboratory based de-arsenification process of drinking water using SORAS:

4.4.1. Selection of materials

Amla, Pomegranate, Apple, Guava, Orange, and Tomato were selected as a natural iron and Vit-C enriched fruit sample, as well as, Bean, Oat, Pumpkin seed, Sesame seed, and

lentils (Green mung daal, Mung daal, Massor daal) were targeted as a natural iron and Vit-C enriched grain samples (**Fig. 10**). The fruit samples were bought from the local markets and washed with double distilled water and cut into pieces with a fresh knife. The seed and grain samples were also bought from the super markets and air dried for 2 to 4 hrs. Then the samples were grinded in a grinder and weighed accurately.

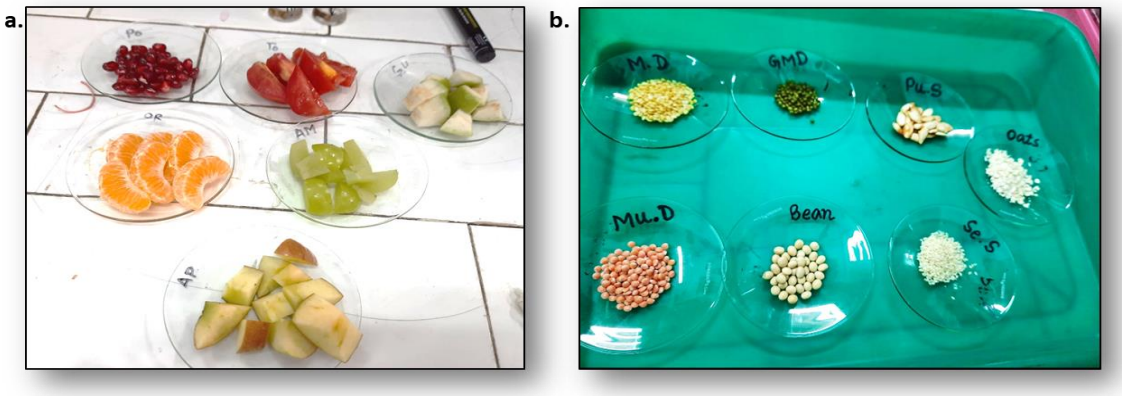


Fig. 10. Selected fruits and seed samples for SORAS

4.4.2. Experimental set up

4.4.2.1. Preparation of fruit juice for SORAS

All the fruit samples were thoroughly cleaned with deionized water to remove any adhering contaminants if present. The samples were air dried and cut into small pieces and then the samples were weighed (100g). Samples were then run in a mixer grinder and fruit juice were prepared. The mesh was strained through a clean and fine cloth. Juice samples were transferred in centrifuge tubes and preserved in refrigerator.

4.4.2.2. Preparation of synthetic As-spiked water

Synthetic test solutions were prepared in the laboratory by using the tap water. Initial iron and As content of tap water were measured. Three different As concentration 100 µg/L, 250 µg/L and 500 µg/L spike water solutions were prepared in 500 ml transparent PET bottles from stock solutions of 1000 µg/L As-III and As-V. Each solutionwas filled in eight separate PET bottles. The solutions were subjected to different dose of amla and pomegranate (10 ml/L and 20 ml/L) and different doses of pumpkin seeds and sesame seeds (2 g/L and 4 g/L) and mixed for 15 minutes with SpinixOrbitalShakerat 120-150 rpm. This mixing provides sufficient oxygen for oxidation of Fe(II) to Fe(III). For preparation of synthetic solutions, tap water was used whose iron concentrations were

estimated to be 0.25 mg/L. The PET bottles were kept in direct sunlight for 4long hours (12 pm to 4 pm) (**Fig. 11**). The experiment was done during winter season (January to February, 2020). Experimental samples were collected twice from the distinct containers through decanting the bottles after 2hr. and 4 hr. respectively. Changes in As and Fe concentration was estimated with different time irradiation. Samples were collected twice after 2 hr. and 4 hr. and preserved with nitric acid and kept in thefreezer at 4 °C to estimate concentrations of As and Fe. pH of the water samples at two times has been evaluated also. A blank experiment had been performed without addition of any external component; only passive sedimentation has been checked.

Fig. 11. Irradiation of experimental water samples in the rooftop of Chemical Engineering building (a) Initial, (b) Final

4.5. Analysis of sample

4.5.1. Chemicals, reagents and instruments

Analytical grade chemicals were used during the present research study and throughout the work double distilled water was used. Each and every chemical used in every analysis of the work including standardization, reagent preparation of physicochemical parameters, digestion and analysis is tabulated here below (**Table 1**). The small and large instruments used for sample processing and analysis for my research are summarized in **Fig. 12** and **Fig. 13**.

Table 1: Chemicals, reagents and instruments used during analysis

Sl. No.	Parameters	Reagents	Methods	Instruments
1	**Location identification**	-	-	GPS meter (Model: GARMIN Etrex 30x GPS) (**Fig. 12a**)
2	**Radiation for Uranium**	-	-	Radiation survey meter (POLIMASTER, model-PM1405) (**Fig. 12b**)
3	**pH**	Standard buffer capsules (pH 4.0, 7.0 and 9.2)	pH metric estimation	Portable pH sensor (Electrode based), (digitalmulti parameter waterproof instrument (HANNA, HI 98194, pH/EC/DO Multipara meter, made in Romania) and (pH meter CL 46+, Toshcon Industries Pvt. Ltd (**Fig. 12c, d**)
4	**Electrical Conductivity (EC) (µS/CM)**	Standard KCl solution	Potentiometric estimation	Portable EC sensor (Electrode based), (digitalmulti parameter waterproof instrument (HANNA, HI 98194, pH/EC/DO Multipara meter, made in Romania) and Conductivity meter-306 (Systronics) **(Fig. 12c, e)**
5	**Total Dissolved Solids (TDS)**	-	Gravimetric estimation	Fine balance (METTLER AE240) (**Fig. 12f**)

6	**Total Suspended Solids (TSS)**	-	Gravimetric estimation	Fine balance (METTLER AE240) (**Fig. 12f)**
7	**Salinity (ppt)**	-	-	Portable Salinity Sensor (Electrode based), (model Hi98194, Hanna Instruments) (**Fig. 12c**)
8	**Oxidation Reduction Potential (ORP) (mV)**		Potentiometric estimation	ORP Tester (Electrode based), (model Hi98194, Hanna Instruments) (**Fig. 12c**)
9	**Temperature (°C)**	-	-	Temperature sensor, model Hi98194, Hanna Instruments and Conductivity meter-306 (Systronics) (**Fig. 12c**)
10	**Turbidity (NTU)**	Hydrazine sulfate (N_2 H_6 SO_4), Hexamethylenetetramine(CH_2)$_6$ N_4	-	Digital turbidity meter, model 331 E (**Fig. 12g**)
11	**Total Hardness as $CaCO_3$ (mg/L)**	(M/100) Ethylenediaminetetraacetic acid (EDTA) or (M/100) disodium-EDTA (Na_2EDTA) solution, ammonium hydroxide-ammonium chloride (NH_4OH-NH4Cl) buffer, Eriochrome Black T (EBT) indicator	Complexometric titration method	
12	**Calcium Hardness as $CaCO_3$ (mg/L)**	(M/100) disodium-EDTA (Na_2EDTA) solution, 10 % NaOH buffer solution and murexide indicator	Complexometric titration method	
13	**Magnesium hardness as $CaCO_3$ (mg/L)**	Theoretically calculated by subtracting Calcium hardness from Total hardness value		
14	**Total alkalinity (TA) (mg/L)**	(N/50) sulfuric acid or H_2SO_4 solution, (N/50) sodium carbonate or	Acid-base titration method	

		Na_2CO_3 solution, methyl orange indicator		
15	**Phenolphthalein alkalinity (PA) (mg/L)**	(N/50) sulfuric acid or H_2SO_4 solution, (N/50) sodium carbonate or Na_2CO_3 solution, phenolphthalein indicator	Acid-base titration method	
16	**Carbonate (CO_3^{2-}), Bicarbonate (HCO_3^-) (mg/L)**	Theoretically estimated from *alkalinity titration*; if PA< ½ × TA, carbonate = 2×PA and [bicarbonate] = (TA-PA)		
17	**Arsenic (As) (µg/L)**	Concentrated nitric acid (HNO_3) (69 %) and hydrogen peroxide H_2O_2 (30% v/v), 0.6% Sodium Borohydride ($NaBH_4$) (Merck, Mumbai, India) in 0.5% sodium hydroxide (NaOH) and 5–10 M hydrochloric acid (HCl) (Merck,Mumbai, India), 10% of potassium iodidesolution (KI) (10% in aqueous solution) and 8% of concentratedhydrochloric acid, stock solution of arsenate (1000 mg/L as arsenic, traceable to SRM from NIST H3AsO4 in HNO_3, 0.5 M) from Merck, Darmstadt, Germany	Atomic absorption spectrophotometry	FI-HG-AAS method, (Hydride generation Atomic Absorption Spectrophotometer (VARIAN AA140) (**Fig. 12h**)
18	**Trace elements**	ICP-OES and AAS (*Outsourcing*)		
19	**Fluoride (F^-) (mg/L)**	Stock solution of fluoride (100 mg/L, Orion 940,911, Thermo Scientific, USA), buffer grade solution, TISAB-III concentrated with CDTA (Total Ionic Strength AdjustingBuffer, Thermo Fisher Scientific, USA, Orion 940911)	Electrode method	Ion Selective Electrode (ISE) meter (Thermo scientific Orion Star A214) combined with fluoride ion selective electrode (Orion ISE model no 9609BNWP) (**Fig. 12i**)
20	**Chloride (Cl^-) (mg/L)**	Silver nitrate or $AgNO_3$ (secondary standard), Potassium chromate (K_2CrO_4) indicator, (M/100) sodium chloride or NaCl solution	Argentometric Titration	

21	**Nitrate (NO_3^-) (mg/L)**	Nitrate as Nitrogen standard 1000 ppm solution, Orion 920707, Thermo Scientific, USA, Nitrate interference suppressor solution, Orion 930710, Thermo Scientific, USA	Electrode method	ISE meter (Thermo Scientific Orion Star A214) coupled with nitrate electrode (SW1-02217) and reference electrode (RS1-11546) (**Fig. 12j**)
22	**Sulphate (SO_4^2) (mg/L)**	Sodium sulphate (Na_2SO_4) stock solution, Conditioning reagent comprising of glycerol, HCl, 95 % ethyl alcohol (C_2H_5OH), NaCl or sodium chloride salt and barium chloride or $BaCl_2$ salt	Nephelometric turbidity method through spectrophotometry	UV-Vis Spectrophotometer (ORION AQUAMATE 8000) (at λ= 420 nm) (**Fig. 12k**)
23	**Iron (Fe) (mg/L)**	Standard stock solution of 1000 mg/L of iron (Fe) (traceable to SRM from NIST, $Fe(NO_3)_3$ in HNO_3 0.5 moL/l) from Merck, Darmstadt, Germany, solution of sodium acetate-acetic acid (CH_3COONa-CH_3COOH) buffer (pH 4–5), 10% hydroxylamine hydrochloride or $HONH_2$ (for reduction of ferric to ferrous solution) and 0.25% of 1,10-phenanthroline solution (Merck, Mumbai, India)	Spectrophotometric estimation	UV-Vis Spectrophotometer (ORION AQUAMATE 8000) (at λ=510 nm) (**Fig. 12k**)
24	**Phosphate (PO_4^{3-}) (mg/L)**	Ammonium molybdate or $(NH4)_6Mo7O_{24}$salt, sulphuric acid or H_2SO_4, glycerol ($C_3H_8O_3$) and stannous chloride ($SnCl_2.2H_2O$)	Molybdate method through Spectrophotometric estimation	UV-Vis Spectrophotometer (ORION AQUAMATE 8000) (at λ= 690 nm) (**Fig. 12k**)
25	**Sodium (Na^+) (mg/L)**	Sodium Chloride (NaCl) salt for standardization	Atomic Emission Spectrophotometry	Flame Photometer (HPGSystem, Chandigarh, model number G-301) (**Fig. 12l**)
26	**Potassium (K^+) (mg/L)**	Potassium Chloride (KCl) salt for standardization	Atomic Emission Spectrophotometry	Flame Photometer (HPGSystem, Chandigarh, model number G-301) (**Fig. 12l**)

27	**Calcium (Ca^{2+}) (mg/L)**	Theoretical calculation from calcium hardness; [Ca^{2+}] = Ca-Hardness/2.50		
28	**Magnesium (Mg^{2+}) (mg/L)**	Theoretical calculation from magnesium hardness; [Mg^{2+}] = Mg-Hardness/4.11		
29	**Uranium (U) (µg/L)**	Polysilicate solution POLISORB Colloidal Silicon Dioxide, sodium hydroxide or NAOH pellets, Nitric acid (HNO_3) solution (Merck, Mumbai, India), uranium (1005 ± 4 µg/ml, uranyl nitrate hexahydrate as standard, 13520-83-7, Inorganic Ventures, USA)	Phosphoroscence	Fluorescence spectroscopy (FLUORAT-02-4M, LUMEX INSTRUMENTS) (at λ=530 nm) (**Fig. 12m**)
30	**Vitamin C or Ascorbic acid**	4 % oxalic acid ($C_2H_2O_4$), sodium bicarbonate ($NaHCO_3$), L-Ascorbic acid (A0278, Sigma-Aldrich), 2,6-Dichloroindophenol (DCIP)	Titration method	
31	**Proline**	L-Proline standard (P0380 Sigma Aldrich), Ortho-phosphoric acid (6 M), phosphoric acid solution (H_3PO_4), Ninhydrin (N7285Sigma-Aldrich), glacial acetic acid, sulfosalicylic acid (S2130 Sigma-Aldrich), toluene (179418Sigma-Aldrich)	Spectrophotometric estimation	UV-Vis Spectrophotometer (ORION AQUAMATE 8000) (at λ=520 nm) (**Fig. 12k**)

Fig. 12. Instruments used to analyze different physico-chemical parameters: (a) GPS meter, (b) Radiation meter, (c) HANNA multiparameter, (d) pH meter, (e) Conductivity meter, (f) Fine balance, (g) Turbidity meter, (h) Atomic Absorption Spectrophotometer, (i) Ion selective electrode for Fluoride, (j) Ion selective electrode for Nitrate, (k) UV-Vis Spectrophotometer, (l) Flame photometer, (m) Fluoroscence spectrophotometer

Fig. 13. Instruments used during sample processing: (a) Rough balance, (b) Magnetic stirrer, (c) Centrifuge, (d) Hot air oven, (e) Vortex, (f) Hot plate, (g) Orbital shaker, (h) Water bath

4.5.2. Accuracy and precision

The worth of all analytical work was maintained through appropriate standardization or calibration of instruments, spiked sample recovery testing and routine blank experiments. For proper calibration of fluoride and nitrate through ion selective electrodes, the slope value should range always within -52.0 mV/dec to -62.0 mV/dec at 25°C. The certified reference materials (CRM) used in relation to estimation of total As were 'Rice flour 1568a' (NBS, Gaithersburg, MD, USA), 'Tomato leaves 1573a' (NIST, Gaithersburg, MD, USA), 'River Sediment 1645' (NBS, Washington, DC 20234) and 'Human hair ERMDB001' (European Commission, JRC, IRMM, Retieseweg, Geel, Belgium). Consecutive acid digestive analysis of As in CRM samples showed 90-94, 100-112, 95-105 and 90-92% recovery in Teflon-bomb digestion against their respective licensed values which are 0.29, 0.112, 0.044 and 66 mg/kg respectively. Analytical precession was also checked for Se and Zn by digestion analysis of SRM 1568a (Rice flour) in 280 FS AA system, which showed a recovery in the range of 90–95% against their respective licensed values (19.4 ± 0.5 μg/g and 0.38 ± 0.04 μg/g). Accuracy and precision of the generated data were validated by inter laboratory testing and duplicate analysis of certain percentage of the samples. The result of inter-laboratory testing of few selected water samples using AAS in my lab (SOES, J.U) and ICP-OES, Agilent India, New Delhi is shown here below (**Table 2**).

Table 2: Inter laboratory testing results of arsenic

Sl. no.	As concentration in AAS (SOES, J.U)	ICP OES, Agilent India, New Delhi	% Variation
1	55.6	42	24.46
2	17.43	15	13.94
3	134	125	6.72
4	142	114	19.72
5	61	50	18.03
6	237	191	19.41
7	56	42	25.00
8	153	103	32.68
9	49	36	26.53
10	212	155	26.88
11	39.6	41	-3.54
12	60.4	43	28.81
13	18.9	15	20.63
14	22.3	16	28.25
15	280	193	31.07
16	44	41	6.82
Average	**-**	**-**	**20.33**

4.6. Data analysis

4.6.1. Water Quality Index (WQI)

A comprehensive depiction on the overall water quality of the studied area is reflected through WQI as given in Gupta and Misra (2018) and Meng et al. (2016).

A specific number (weight) are allotted to the parameters according to their significance in water quality followed by 'relative weight' (W_i) calculation for computation of WQI.

$$\mathrm{Wi} = \frac{\mathrm{wi}}{\sum_{i=1}^{n} \mathrm{wi}} \qquad \text{eq. (1)}$$

'Wi'is relative weight, 'wi'is weight of each parameter, 'n'is number of parameters.

A quality rating scale (qi) is made next for every parameter by dividing their concentration in each sample by their corresponding standard as per the recommendations by BIS and the result is multiplied by 100.

$$qi = \frac{Ci}{Si} \times 100 \qquad \text{eq. (2)}$$

'qi' is quality rating, 'Ci' is concentration of chemical parameters in each sample (mg/L), 'Si' is drinking water standard in India for each chemical parameter as per the guiding principle of BIS (2012).

To calculate WQI, the SI is first estimated for each parameter and used to define the WQI according to the next equation,

$$SIi = Wi \times qi \qquad \text{eq. (3)}$$

$$WQI = \sum SIi \qquad \text{eq. (4)}$$

Where, SIi= subindex of i^{th} parameter; q_i = the rating based on concentrate on of i^{th}parameter (Anim-Gyampoa et al., 2019).

Lastly, the calculated WQI values are categorized into 5 types of water quality: from excellent to inapt for drinking.

4.6.2. Environment quality assessment

4.6.2.1. Single factor pollution index study for As and Fe

A single factor pollution index (Ii) assessment has been done to evaluate the groundwater contamination with special reference to As and Fe following the equation (Zhang et al., 2018),

$$Ii = Ci/Co \qquad \text{eq. (5)}$$

Where, Ii stands for specific indices, Ci represents the concentration of the toxic component in groundwater and Co refers to the permissible concentration of that contaminant set by the standard guidelines. Respective Co values of drinking water As and Fe are 10 µg/L and 0.3 mg/L (BIS, 2012; WHO, 2011a) and the values for irrigational water are 100 µg/L and 5.0 mg/L, respectively (FAO, 1985; Fipps, 2003). Groundwater contamination factors are classified in 4 groups; low contamination (Ii <1.0), moderate contamination (1.0 ≤ Ii <3.0), considerable contamination (3.0 ≤ Ii <6.0) and high contamination (Ii ≥ 6.0) (Hakanson, 1980; Rehman et al., 2018).

4.6.2.2. Nemerow pollution index (NPI) study

NPI or 'Nemerow Pollution Index' is a comprehensive study of indexing pollution based on the single factor pollution index. It is used to assess the water quality of different sampling sites which simultaneously highlights the importance of several metals in groundwater (Bodrud-Doza et al., 2019; Yan et al., 2015; Zhong et al., 2015). The equation is:

$$\text{NPI} = \sqrt{\left[\frac{\left\{\left(\frac{1}{m}\right)\sum_{i=1}^{m}(\text{Ii})^2 + Iimax^2\right\}}{2}\right]} \qquad \text{eq. (6)}$$

Where, m is number of metals in the study, Iiis the single factor contaminationindex and Iimax is the highest value of the single pollution indices among all the pollutants considered in study. The classification of NPI values for reference water quality is given below. Clean water (NPI < 0.7; class I), slightly polluted water (0.7 ≤ NPI < 1.0; Class II), moderately polluted water (1.0 ≤ NPI < 2.0; Class III), heavily polluted water (2.0 ≤ NPI <3.0; Class IV), severely polluted water (≥ 3.0; Class V) (Nemerow, 1974).

4.6.2.3. Ecological risk analysis

The potential ecological risk assessment evaluates the potential impact of metals on ecosystems (Egbueri, 2020a, b). A thematic work by Hakanson (1980) stated that

ecological risk is dependent on heavy metal concentrations in any system, type of contaminant and strength of toxicity. It is calculated by,

$$Er = Tr \times I \quad \text{eq. (7)}$$

Where, Er is denoted as calculated ecological risk; Tr and I signify toxicity response coefficient andthe contamination factor, respectively. For As, the toxicity coefficient is 10. The quantitative estimation of ecological risk is generally categorized into 5 classes; low risk (Er< 40), moderate risk (40 ≤ Er< 80), considerable risk (80 ≤ Er< 160), high risk (160 ≤ Er< 320) and very high risk (Er≥ 320).

4.6.2.4. Heavy metal evaluation index (HEI) study

It determines the comprehensive water quality with respect to heavy metals. It is nothing but the summation of the single factor contamination indexes for all the water samples. In the present study, HEI is performed based on the involvement of As, Fe, Cu, Mn, Cr, Cd, Pb and Hg.

$$HEI = \sum Hi \quad \text{eq. (8)}$$

Where, $H_i = H_c/H_{mac}$ eq. (9)

H_c = the monitored value, H_{mac}= maximum admissible concentration (MAC) of the ith parameter. Classification of the HEI values are done in three groups based on the multiple of the mean to segregate between various contaminations levels such as: 'Low (HEI <8)', 'Medium (8< HEI < 16)' and 'High (HEI > 16)' (Edet and Offiong, 2002).

4.6.2.5. Health risk assessment

Health risk is categorized into two classes, cancer and non-cancer. Non-carcinogenic risk is calculated through 'Hazard Quotient' (HQ), aproportion between 'Lifetime Average Daily Dose' (LADD) and 'Reference Dose' (RfD). Cancer risk is determined through multiplication of Average Daily Dose (ADD) and cancer slope factor (CSF).

$$\mathrm{HQ} = \frac{\mathrm{LADD}}{\mathrm{RfD}} \quad \text{eq. (10)}$$

$$\mathrm{LADD} = \frac{\mathrm{C \times IR \times EF \times ED}}{\mathrm{BW \times AT}} \quad \text{eq. (11)}$$

Where,

IR is the Daily Ingestion Rate of drinking water which is considered 2.5 L/day (USEPA, 2014)

EF is the Exposure Frequency which is 365 days/year.

ED is the Exposure duration (age), considered 65 years for adult (WHO, 2011a)

BW is standard Body Weight, 70 kg is for an adult (Goswami et al., 2019; USEPA, 1986)

AT is Average Lifetime (23725 days)

Cancer risk (CR) calculation for As is done following the equation:

$$CR = LADD \times CSF \quad \text{eq. (12)}$$

Where, CSF is Cancer Slope Factor =1.5 per mg/kg/day for As (USEPA, 2005)

Cancer risk for Uranium is measured by the formula:

$$CR = Cu \times (Rcoeff \times IR \times EF \times AT) \quad \text{eq. (13)}$$

Where,C_u is the concentration of U (Bq/L); U (Bq/L) = U (µg/L)× C.F

C.F is conversion factor (1 µg/L = 0.02528 Bq/L) (Sharma and Singh, 2016)

R_{coeff} is Risk coefficient i.e. 1.19×10-9 /Bq (Saini et al., 2016; USEPA, 2000)

4.6.2.6. Risk thermometer (Severity Adjusted Margin of Exposure or SAMOE)

Human health risk is also assessed with application of 'Risk thermometer' model set by the Swedish National Food Agency. The MoE or 'margin of exposure' is the difference between TDI (tolerable daily intake) and 'exposure' (AgriAs 2017; Sand et al., 2015a, b). The calculated 'exposure' of anyconstituent through diet is compared with their health-based reference point (RP) or TDI. Finally, the order of human health risk from the foodstuffs is estimated with the following equations.

$$SAMOE = TDI/(AF_{BMR} * AF * SF * E) \quad \text{eq. (14)}$$

According to WHO (2011b),

TDI is Tolerable Daily Intake value of As in human (3.0 µg/kg body wt/day) on the basis of range of 2 to 7 µg/kg body wt/day for the assessed total dietary exposure.

AF_{BMR}= 1/10, when a substance's effect is assumed in the range $BMD_{0.5}$ - BMD_{10} (BMD= Benchmark Dose).

AF (Assessment factor) is 10, since human is sensitive under As exposure

SF (Severity factor) is 100, when the effect of Asisbothgenotoxic and carcinogenic.

E (Exposure) is various exposure factors; the exposure in food by any toxic element is equated.

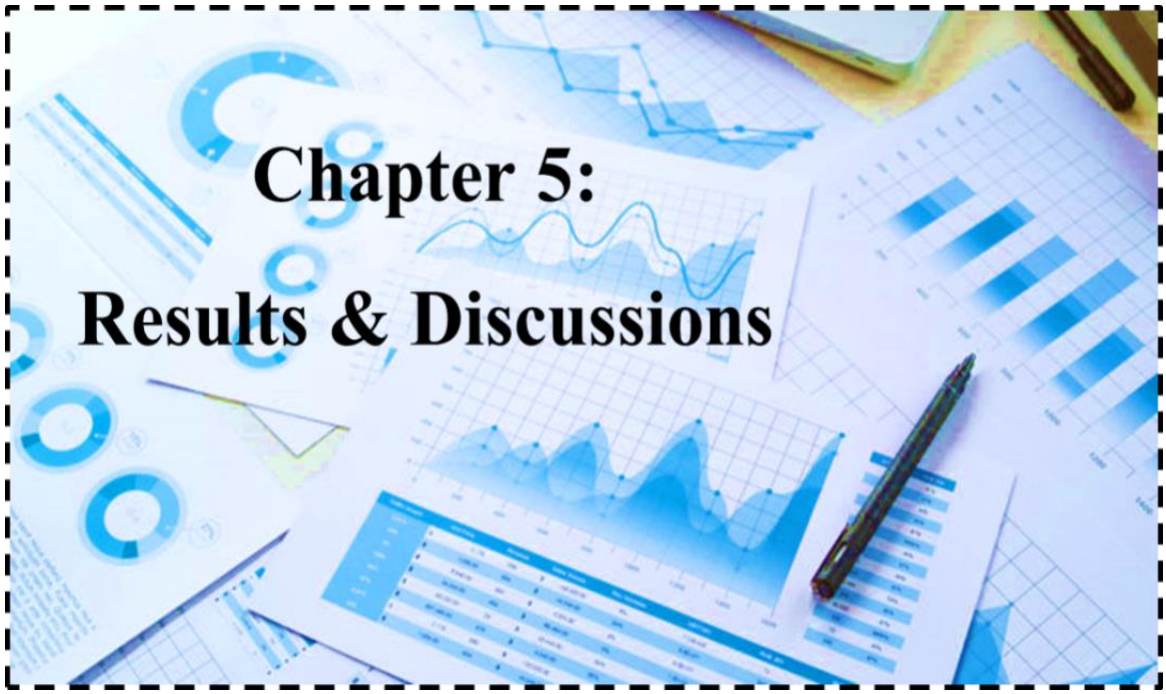
As suggested by Sand et al. (2015a), the calculated health risk values from daily diet iscategorized into five classes; SAMOE values from >10 to < 0.01 (absolutely 'no' to 'high') (**Table 3**).

Table 3: Risk categories according to the Severity Adjusted Margin of Exposure (SAMOE)

Risk Class	Risk Level	SAMOE
Class 1	Nil/no	>10
Class 2	No to low	1-10
Class 3	Low to moderate	0.1-1
Class 4	Moderate to high	0.01-0.1
Class 5	High	<0.01

4.7. Statistical analysis

For the entire research study, calculations, hypothesis testing and several statistical interpretations were required and they have been performed using Excel 2016 (Microsoft Office), Origin 2017, PAST (version 4.09), R version 3.6.3[2], Oracle Crystal Ball. The sampling map has prepared with the help of Google Earth Pro software.


Chapter 5:
Results & Discussions

5.1. Groundwater quality evaluation for drinking and domestic purposes

5.1. 1. Nadia

5.1.1. 1. Status of drinking water quality parameters

Evaluation of 20 physico-chemical parameters on 110 groundwater samples has been done for estimation of the water quality of Nadia for drinking and domestic purposes. The normal statistics of the water quality parameters are shown in **Table 4** based on three replicates. The average pH value 7.54 makes the water quality of the district little alkaline which is confirmed by the average total alkalinity value (375 mg/L). The temperature ranges from 25.7 to 31.2°C with a mean of 28.2 °C and the ORP value ranges between 103 and 248 mV.

Table 4: Normal statistics of the water quality parameters in Nadia district

Parameters*	Min	Max	A.M	S. D	C.V.	Q1	Median	Q3
pH	6.84	8.18	7.54	0.25	3.32	7.36	7.5	7.72
TDS (mg/L)	270	1507	525	198	37.7	418	477	540
Temperature (°C)	25.7	31.2	28.2	3.0	10.6	27.1	28.2	28.8
EC (µS/cm)	404	2250	788	297	37.7	624	713	823
ORP (mV)	103	248	160	33.5	21	131	155	189
Radiation (nSv)	0	350	151	62.9	2.4	105	150	198
Fluoride (mg/L)	0.01	0.71	0.18	0.15	83.3	0.05	0.16	0.24
Chloride (mg/L)	14	347	78.2	52.1	66.6	42.6	63.2	94.7
Nitrate (mg/L)	0.5	57.6	10.1	12.5	124	1.94	4.87	13.7
Sulphate (mg/L)	1	200	26	26.9	103	10.7	16.7	33.5
Phosphate (mg/L)	0.05	19.2	1.36	2.6	191	0.16	0.45	1.32
Uranium (µg/L)	0.21	20.9	3.88	4.55	117.3	1.36	1.64	4.57
Total hardness as $CaCO_3$ (mg/L)	20	570	329	119	36.17	270	320	400
Total Alkalinity (mg/L)	186	725	375	77	20.5	330	361	415
Carbonate (mg/L)	0	229	56	42.2	75.4	31.6	42.4	68
Bicarbonate (mg/L)	37	650	319	87	27.3	266	307	354
Arsenic (µg/L)	3	206	22	30	136	3	10.8	31.5
Iron (mg/L)	0.01	13.7	3.02	2.9	96	0.65	2.14	4.75
Calcium (mg/L)	4	156	53.7	32.9	61.3	26.4	51.2	72
Magnesium (mg/L)	0.48	114	44.9	27.0	60.1	24	45.7	61.5

*Water quality parameters of all the samples have been analysed based on three replicas

Min: Minimum, Max: Maximum, AM: Arithmetic Mean, SD: Standard Deviation, CV: Coefficient Variation, Q1: Median of the lower half, Q3: Median of the Upper half, Med: Median

EC and TDS help to apprehend the total concentration of the soluble salts in groundwater. While TDS expresses the number of inorganic salts comprising organic matter in water, EC estimates the electrical current present in water. Rusydi (2018) stated that these

parameters own a solid relationship between themselves. The mean EC value is 788µS/cm and the range is 404 to 2250 µS/cm, which helps to realize that water holds decent potential for minerals and salts. The average TDS is 525 mg/L, slightly superior to the acceptable value of TDS in drinking water. It also found a quite strong relation between EC and TDS, which is narrated through the regression analysis (**Fig. 14a**). The mathematical relation between TDS and EC is researched by several personals so that one parameter can be estimated simply from the other. Though, the relationship is not every time linear, it is articulated through,

$$(TDS = k \times EC) \qquad \text{eq. (15)}$$

The factor 'k' is TDS/EC ratio. The range of different 'k' values is 0.55 to 0.75 for different kinds of water samples according to the report of Rusydi (2018). Similar type of ratio between 0.5 and 1.00 in freshwater samples was observed by McNeil and Cox (2000). EC and TDS have been assessed using a probe in the present work and the proportion also lays in the range between 0.52 and 0.67. Chloride ions impact TDS which gives the idea of salinity in groundwater (Balakrishnan et al., 2011; Nelson, 2002). A quite good association is found between TDS and Cl^- (**Fig. 14b**) where the mean Cl^- value is 78.2 mg/L. The regression statistics is performed bearing in mind TDS as 'dependent variable' and EC, Cl^- are termed as 'independent' variables.

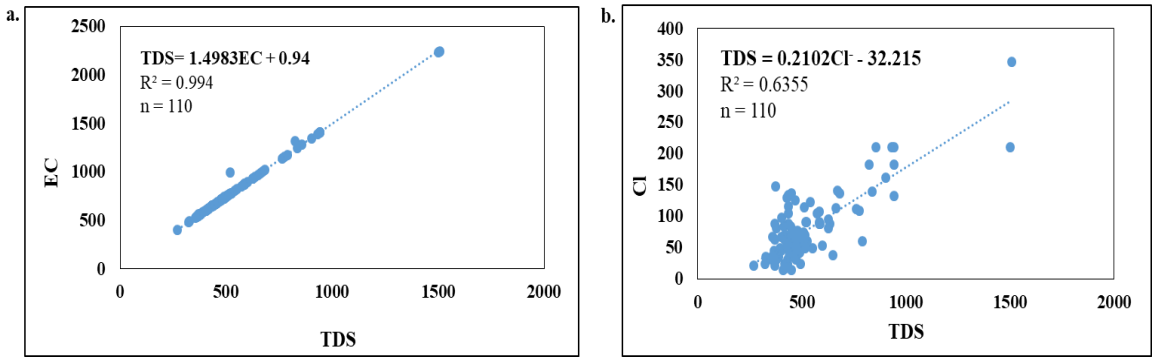


Fig. 14. Regression analysis between (a) TDS and EC, (b) TDS and Cl^-

The mean Fe concentration is 3.02 mg/L with a range between 0.1 and 13.7 mg/L considering 110 samples collected from all the 17 blocks, which specifies that the groundwater is iron enriched. The average of Total Hardness (TH) and Total Alkalinity (TA) values are found to be more than their respective recommendation values in drinking water. The mean TH concentration is found to be 329 mg/L with a range between 20 and 570 mg/L which indicates that the quality of groundwater is hard. The presence of calcium,

magnesium, iron, carbonate or bicarbonate ions signify the hardness of water. The range of calcium and magnesium ion concentrations are 4 - 156 mg/L and 0.18 - 114 mg/L. The mean Ca^{2+} concentration is 53.7 mg/L, higher than that of Mg^{2+} ion (44.9 mg/L). The mean carbonate concentration (56 mg/L) is less than bicarbonate concentration (319 mg/L). The mean SO_4^{2-}concentration is found to be 26 mg/L (range: 1 - 200 mg/L), and mean PO_4^{3-} concentration is 1.36 mg/L (range: 0.05-19.2 mg/L). Thus, the order of the mean anionic concentration is HCO_3^-> Cl^-> CO_3^{2-}> SO_4^{2-}> NO_3^-> PO_4^{3-} that signifies the water quality of Nadia is alkaline and the higher HCO_3^- concentration has a likelihood of dissolution of CO_3^{2-} minera ls in groundwater. The mean U concentration is observed as 3.88 µg/L with a range between 0.21 and 20.9 µg/L. Theamount of U in groundwater relie s on the oxidation state of U in which it exists in the bedrock which also varies with water pH and ORP (Waseem et al., 2015). The mobility of U in the aquifers is relieved with the oxidizing nature of the groundwater i.e. as high the ORP is, higher the propensity of U mobilization is into deep aquifers (Smedley et al., 2006; Yang et al., 2014). The mean As concentration is 22 µg/L (range: 3-206 µg/L), while the mean NO_3^- concentration is 10.1 mg/L (range: 0.5 - 57.6 mg/L).

5.1.1.2. Scattering of the main toxic elements (As, NO_3^-, U, F^-)

Numerous health hazards concerning arsenicosis and skin pigmentation is caused by over-exposure to As through drinking water that may lead to cancer in different human organs (Abdul et al., 2015; Bhowmick et al., 2018). Over-exposure to nitrate, one of the most significant contaminants in groundwater in the world, causes a risk of infant cyanosis or blue baby syndrome (Fan and Steinberg, 1996). Consumption of elevated F-containing water consequences in decaying of bones, dental and skeletal fluorosis in adults and children (Narsimha and Rajitha, 2018). Uranium induced toxicity causes nephritis in human and shows numerous detrimental effects on the renal system if ingested for a longer period through water. Its radioactive properties are more adverse than its chemical properties (Ajay et al., 2016; WHO, 2012).

Spatial distribution of these four toxic components is presented through contour diagrams where a contour line is marked as a constant value-function of two variables in a curve that joins equal valued points (**Fig.15**).

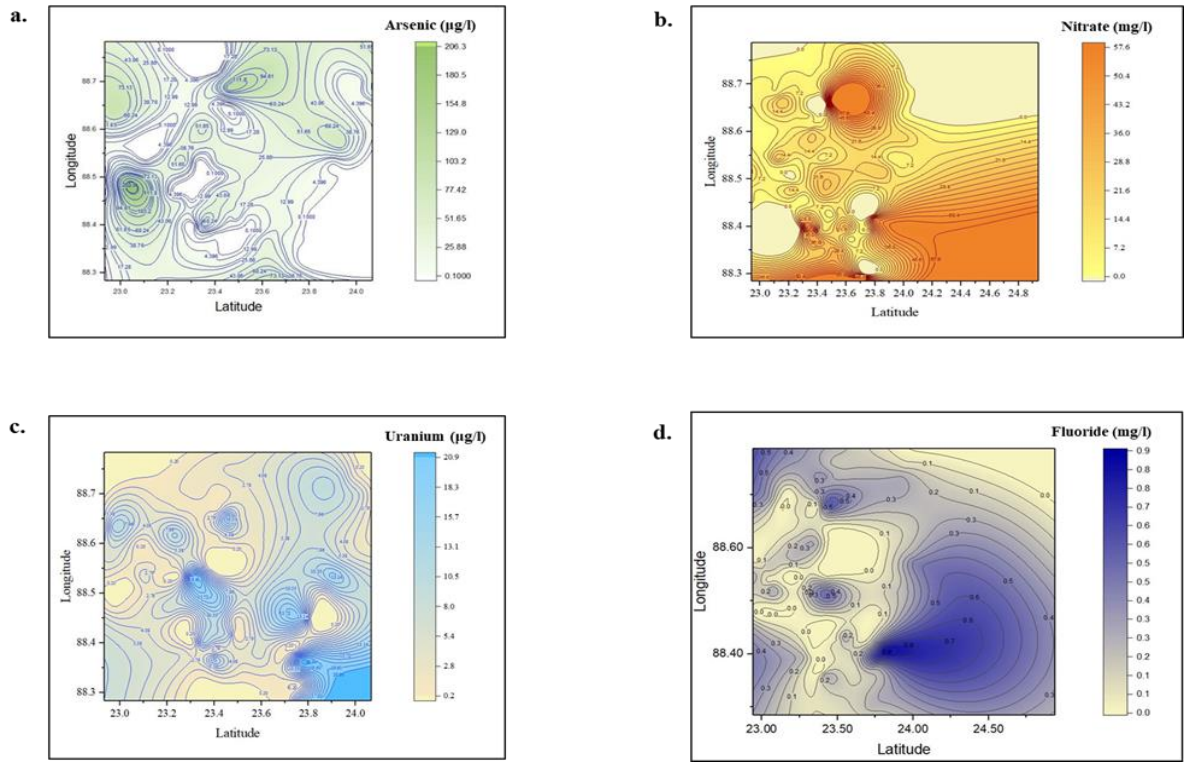


Fig. 15. Distribution (Contour diagram) of four toxic chemical parameters in groundwater: a. Arsenic, b. Nitrate, c. Uranium and d. Fluoride

The contours deduce the relative gradient of a parameter and evaluate the parameters at specific places. The concentration of the components in groundwater seems high where the contour lines are closed, and the distant contour lines designate low concentration of the elements. Where the contour lines are converging, the highest concentration of the elements is detected. The different intensities color is to identify the concentration range of the toxic parameters (Nag and Ghosh, 2013). The dissemination of the toxic components in Nadia is depicted in **Table 5**.

Table 5: Block-wise statistical distribution of the four toxic chemicals in groundwater of studied area

No. of samples in each block	Statistical Parameter	Name of Chemical Parameter			
		As (µg/L)	NO_3^- (mg/L)	F^- (mg/L)	U (µg/L)
Haringhata, n = 6	Mean	19.9	4.85	0.1	2.07
	Range	3-76.2	1.76-15.1	0.025-0.23	0.35-4.44
	SD	28.9	5.27	0.077	1.63
Chakdah, n = 6	Mean	51.6	2.92	0.12	0.73
	Range	3-206	1.8-5.72	0.01-0.27	0.34-1.7
	SD	78.1	1.44	0.10	0.49
Shantipur, n = 6	Mean	7.87	4	0.05	3.73
	Range	3-26	0.1-16	0.01-0.19	1.24-6.5

	SD	10.2	6.86	0.08	2.54
Nabadwip, n = 7	Mean	16.5	26.5	0.08	3.74
	Range	3-72.3	1.86-53.4	0.01-0.31	1.4-10.6
	SD	26.2	16.5	0.12	3.26
Krishnanagar-I, n= 6	Mean	14	15.1	0.02	2.78
	Range	3-31.5	0.1-45	0.01-0.06	0.21-10.2
	SD	10.7	18.9	0.02	3.69
Krishnanagar-II, n = 6	Mean	15.6	7	0.05	4.86
	Range	3-43.4	0.24-23	0.02-0.12	0.34-11
	SD	14.8	8.63	0.04	4.43
Ranaghat I, n = 6	Mean	20.2	12.4	0.04	1.89
	Range	3-58.5	6.2-21.6	0.01-0.18	1.23-4.57
	SD	22.1	5.15	0.07	1.32
Ranaghat II, n = 6	Mean	34.3	8.91	0.06	3.56
	Range	5.81-92	4.42-21.7	0.01-0.13	1.08-9.8
	SD	31.7	6.44	0.56	3.3
Hanskhali, n = 6	Mean	18.3	7.3	0.21	4.81
	Range	3-36.4	0.1-17.7	0.1-0.32	1.3-20.5
	SD	13.7	6.18	0.08	7.7
Krishnaganj, n = 10	Mean	31.5	4.7	0.27	1.88
	Range	4.5-117	0.1-6.65	0.1-0.54	1.2-3.92
	SD	32.9	1.85	0.12	0.87
Karimpur I, n = 6	Mean	20.5	1.98	0.2	4.69
	Range	3-41.5	0.1-7.07	0.16-0.23	1.54-9.82
	SD	16.4	2.98	0.03	3.19
Karimpur II, n = 8	Mean	16.9	13.7	0.26	7.69
	Range	3-69	4.2-22.2	0.11-0.59	1.9-13.8
	SD	23.5	7.23	0.19	4.35
Chapra, n = 7	Mean	10.1	10.4	0.31	4.15
	Range	4-37.8	0.2-57.6	0.07-0.62	1.3-13.6
	SD	12.5	20.9	0.22	12.5
Tehatta I, n = 7	Mean	14.6	5.68	0.2	6.45
	Range	3-45.5	0.5-24.6	0.12-0.32	1.12-20.6
	SD	15.2	8.54	0.07	8.78
Tehatta II, n = 6	Mean	10.3	9.92	0.43	6.80
	Range	3-16.9	0.15-39	0.1-0.71	1.27-20.9
	SD	4.95	16.2	0.24	8.17
Nakashipara, n = 5	Mean	23.9	11.1	0.2	2.5
	Range	3-56.4	1.3-27.6	0.1-0.32	1.08-4.73
	SD	21.7	10.3	0.09	1.62
Kaliganj, n = 6	Mean	43.5	21.8	0.2	2.9
	Range	3-106	0.32-54	0.15-0.26	0.57-11.9
	SD	33.9	23	0.04	4.44

The groundwater in all the 17 blocks is contaminated with As and ~ 52% samples contain As beyond its acceptable limit in drinking water, 10 µg/L (WHO, 2011a). The highest concentration of As is found in the block Chakdah (Lat: 23°02´44.56´´, Long: 88°29´31.23´´), 206 µg/L (Fig.15a) which is nearly 20 times higher than the suggested value. The As contamination in the Chakdah block has further been studied in detail and shown in **Fig. 16**. Drinking water samples (n= 62) considering the 8 gram panchayats out of total 10 have been evaluated where it has been seen that the average As concentration in the groundwater samples was 32.4 µg/L (range: <3 to 219 µg/L , n= 44) and Silinda I owns the approximately highest percentage of samples above 10 µg/L As concentration. The drinking water samples being mainly used by school students (tube-wells) collected from school premises, contained an average As concentration of 60 µg/L; almost 46.8% sample showed As concentration > 10 µg/L (n= 22 out of 47). Besides Chakdah, in another community development block of Nadia district named Haringhata, about 55% of collected samples from school premises showed As concentration > 10 µg/L (average: 91.8 µg/L).

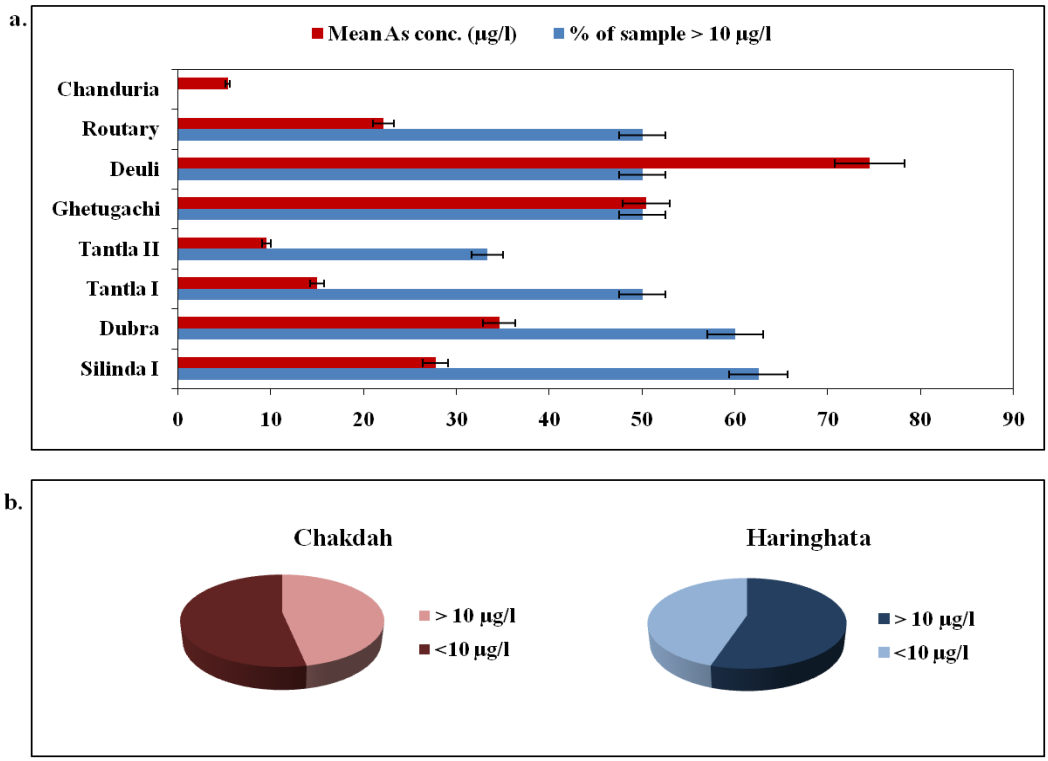


Fig. 16. Groundwater arsenic contamination in Chakdah and Haringhata

A highest concentration of NO_3^- is observed in one sample from the Chapra block (Lat: 23°29´09.68´´, Long: 88°39´43.40´´) (57.6 mg/L) (Fig.15b) where the permitted limit of NO_3^- is 45 mg/L (BIS, 2012). Concentration of NO_3^- in drinking water has crossed the

recommended limit in a few groundwater samples in three other blocks (Nabadwip, Krishnanagar-I and Kaliganj) with respective values of 53.2, 45 and 54 mg/L. The higher NO_3^- concentrations in groundwater may be an outcome of excess usage of NO_3^- fertilizer in agriculture. The U concentration (0.21-20.9 µg/L) ranges within the acceptable limit by the World Health Organization and Atomic Energy Regulatory Board, Department of Atomic Energy, Govt. of India (30 µg/L and 60 µg/L) (AERB, 2004; WHO, 2011a). The maximum U concentration (20.9 µg/L) was found in Tehatta II block (Lat: 23°46´19.75´´, Long: 88°22´43.01´´), shown in Fig. 15c. 1.5 mg/L is the standard value of F^- concentration in drinking water (WHO, 2011a). Fluoride contamination is not protuberant as it ranges between 0.01 and 0.71 mg/L with a mean value of 0.18 mg/L (Fig. 15d). But risk comes from the occurrence of reasonably high NO_3^- concentration in few places, which may provoke the solubilization of underlying U from the minerals in the proximate future as NO_3^- plays a dynamic part in secondary U dissolution. Groundwater withdrawal is a recurrent occurrence in our state. All over the study area and other parts of Bengal, there are many shallow and deep tube-wells that draw groundwater unremittingly for drinking, cooking and other domiciliary purposes and for irrigation (Biswas et al., 2019; Chowdhury et al., 2018a, 2018b). So, the most probable mode for desorption and agility of even low amount of U in groundwater is air-oxidation. The carrier elements (HCO_3^- or NO_3^-) catalyze the U mobilization (Nolan and Weber, 2015; Bajwa et al., 2017). The groundwater quality and portability is regulated by several geological contaminants. Groundwater holds a natural U concentration in the range between 0.1 to 10 µg/L under definite conditions, reported in Osmond (1980). Nolan and Weber (2015) revealed that U concentrations were interrelated to NO_3^- in almost 78% of the aquifers from High Plains and Central Valley, U.S.

5.1.1.3. Correlation study among the parameters

Uranium makes a moderate positive correlation with TDS ($r= 0.42$, $p <0.05$) and EC ($r= 0.43$, $p <0.05$), whereas, a small positive correlation with NO3- ($r= 0.27$, $p <0.05$) (**Fig. 17**).

	pH	TDS	EC	ORP	Salinity	DO	F^-	Cl^-	NO_3^-	$SO4^{2-}$	PO_4^{3-}	U	TH	Ca	Mg	TA	$(CO_3)^{2-}$	$(HCO_3)^-$	As	Fe
pH	1																			
TDS	-0.16	1																		
EC	-0.16	**1.00**	1																	
ORP	-0.12	0.17	0.17	1																
Salinity	-0.08	0.26	0.27	0.08	1															
DO	-0.20	0.07	0.06	-0.01	-0.08	1														
F^-	0.33	0.09	0.08	-0.04	0.06	-0.13	1													
Cl^-	-0.10	**0.80**	**0.80**	0.23	0.28	0.13	0.06	1												
NO_3^-	-0.22	0.42	0.42	-0.05	0.16	0.01	-0.05	0.40	1											
$SO4^{2-}$	-0.09	0.45	0.45	0.17	0.21	-0.13	0.00	0.44	0.29	1										
PO_4^{3-}	-0.09	0.09	0.09	-0.06	0.11	0.11	-0.07	0.09	0.11	0.02	1									
U	-0.09	0.42	0.43	-0.04	0.09	0.00	0.15	0.36	0.27	0.33	-0.02	1								
TH	-0.21	0.36	0.35	0.08	0.08	0.20	0.05	0.29	0.31	0.16	0.09	0.12	1							
Ca	-0.16	0.02	0.01	0.06	-0.09	-0.02	-0.16	-0.06	-0.07	-0.03	0.00	-0.15	0.30	1						
Mg	-0.08	0.33	0.33	0.06	0.12	0.17	0.21	0.33	0.21	0.18	0.09	0.21	0.73	-0.31	1					
TA	0.03	0.48	0.48	0.11	0.13	-0.21	0.14	0.42	0.09	0.15	0.00	0.20	0.09	-0.10	0.18	1				
$(CO_3)^{2-}$	0.40	-0.09	-0.06	-0.01	0.14	-0.18	0.14	0.08	-0.15	0.13	-0.05	-0.07	-0.08	-0.22	0.14	0.29	1			
$(HCO_3)^-$	-0.21	0.45	0.44	0.10	0.02	-0.11	0.07	0.31	0.15	0.07	0.03	0.22	0.15	0.08	0.08	0.75	-0.23	1		
As	0.08	-0.09	-0.09	0.05	-0.08	0.08	-0.14	-0.01	-0.07	0.00	-0.01	-0.13	-0.06	0.08	-0.14	-0.14	-0.06	-0.10	1	
Fe	-0.07	-0.02	-0.03	0.21	0.09	-0.03	-0.01	-0.02	-0.13	-0.06	-0.05	-0.06	0.14	0.01	0.06	0.01	-0.12	0.09	0.21	1

Fig. 17. Correlation matrix of the physicochemical parameters in Nadia

There is found a good connotation of NO_3^- with TDS (r= 0.42, p <0.05). It shadows that NO_3^- might play a tiny but important part in U dissolution (oxidization of U-VI) (Nolan and Weber, 2015). Henceforth, it can be said that there is a chance of uranium deposition in groundwater at minimum level from the minerals like uranyl nitrate ($UO_2(NO_3)_2$). According to Waseem et al. (2015), the kinesis of U(VI) is linked to the solubility of a varied variety of uranyl (UO_2^{2+}) minerals. Furthermore, U(VI) generally makes a pH-dependent group of uranyl carbonate and several hydrated complexes in groundwater solutions. Co-occurrence and correlation between U and NO_3^- is again recognized through the multiple axis plot (Fig. 17), using multivariate statistical procedure, which supports their spatial and temporal patterns. Also, Singh et al. (2003) informed that U is known for a greater kinship towards Cl^- ions, observed in groundwater of Amritsar, Punjab, India. There is also a minor but significant positive correlation between $Cl^-$and U (r = 0.36, p < 0.05). It thus be said that Cl^- and NO_3^- might be the carrier of Uranium. In agreement with Bajwa et al., (2017), HCO_3^- ions are good leaching agent for U from the soil sediments. Therefore, a trivial but positive correlation coefficient between U and HCO_3^- in the current study area (r = 0.22, p <0.05) is an evidence that the HCO_3^- produced in the groundwater is from suspension of atmospheric CO_2 or carbonic acid and calcium carbonate reactions which possibly percolates through the soil which then increases U leaching probability (Bajwa et al., 2017). The distribution pattern of the major ions in groundwater and their correlation variation with U may be an outcome of the depth of the shallow or deep aquifers (Shin et al., 2016). More to it, according to Zhou and Gu (2005), the alkaline

groundwater of the study area might be another key cause behind regular U dissolution because alkalinity escalates the desorption of U from metal oxides and hydroxides. SO_4^{2-} ion sensibly correlates well with EC (r = 0.45), TDS (r = 0.45), Cl^- (r = 0.44) and NO_3^- (r = 0.29); that signifies the interrelation among the ions into groundwater. The mild positive correlation between SO_4^{2-} and U (r = 0.33, p <0.05) denotes that. SO_4^{2-} ions are another complexing agent for uranyl ions in groundwater (Almeida et al., 2004). As this complexation is leading in aerated water, it can be exemplified that the sub-lining water-rock system is oxidized for recurrent groundwater extraction from place to place. Bicarbonate and total alkalinity have a sturdy positive correlation (r = 0.75, p <0.05) that says the groundwater alkalinity is mostly backed by HCO_3^- ions. (Mg^{2+}-TDS) and (Mg^{2+} - EC) both have similar positive correlation (r = 0.33, p <0.05) that shows that Mg^{2+} ions have a fitted contribution to the ionic conductance of the present groundwater. No such important correlation has been observed for As and F^- related to other cations and anions.

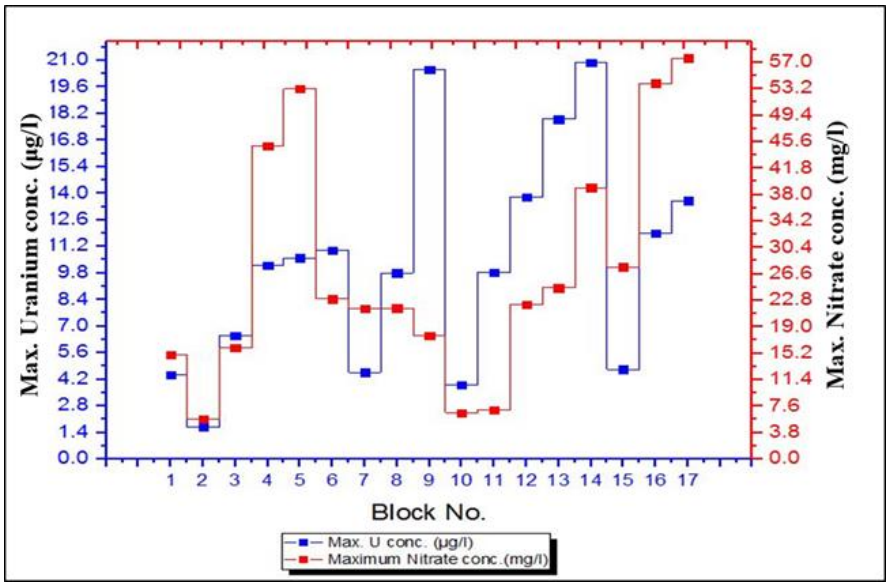


Fig. 18. Co-occurrence of maximum Uranium and Nitrate concentration in groundwater of Nadia through bi-plot diagram

Fig. 18 elucidates that in the block nos. 1, 2, 3 and 10 (Haringhata, Shantipur, Chakdah, and Krishnaganj), co-existence of NO_3^- and U is visible as both the points for highest concentrations are narrowly placed. The block nos. 6, 7, 8 and 15 (Krishnaganj-II, Ranaghat-I, II and Tehatta-II) show the probable co-occurrence of NO_3^- and U in less extent than the previous blocks as the space between the points rises. In consonance with Coyte et al. (2019) and Nolan and Weber (2015), the co-existence of NO_3^- and U is described by the principle that infiltration of NO_3^-enriched water from cultivated fields

backs the mobilization of geo-genic pollutant like U by oxidative dissolution of reduced U(IV) minerals.

5.1.1.4. Cluster Analysis (CA)

The relationship between the clusters is formed by the conforming studies of the studied parameters, presented through dendrogram. It establishes the difference of homogeneousness between the produced clusters as done in Das et al. (2018), Sultana et al. (2014) etc. The hierarchical cluster analysis here proposes that pH and temperature depend on each other directly (**Fig. 19**). TDS-EC comes under one single cluster which shows their stout inter-relation, while both TDS and EC are linked to Cl^- and SO_4^{2-} ions. Between $Ca^{2+}$and Mg^{2+} ions, Mg^{2+} ions concentrations override TH and SO_4^{2-} ion concentrations overrule TA as they are under different small clusters. Also, Total Alkalinity is indirectly linked to HCO_3^- and Cl^- ions. Fe concentration is allied to ORP. Uranium distribution is also directly reliant on HCO_3^- concentration.

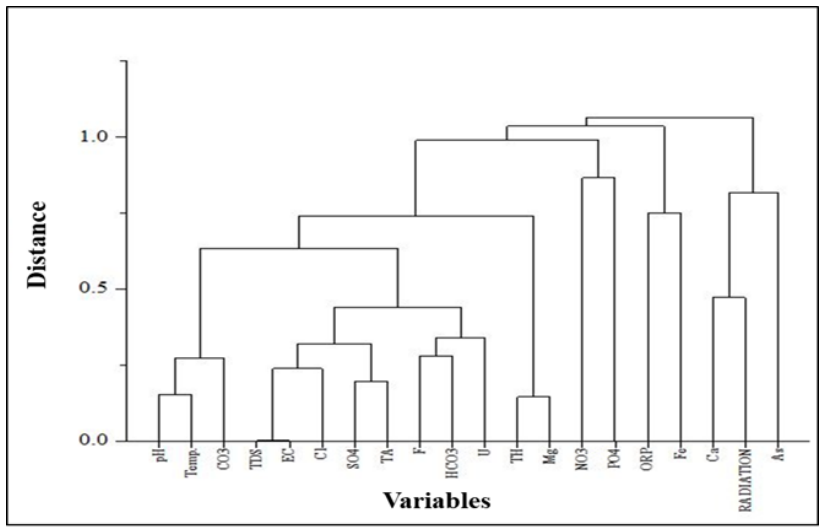


Fig. 19. Dendrogram plot made on water quality of Nadia district

5.1.1.5. Assessment of Water Quality Index (WQI)

WQI has been performed through eq. (1) to eq. (4) where 11 parameters are given a weight (wi) in relation to their importance in water quality for the purpose of drinking. The most toxic As and U are allotted with maximum weight (5) because of its adverse potential while F^- and NO_3^- are allotted with weight 4 because of its status in groundwater contamination. The parameters with their relative weights and acceptable borders in drinking water are tabulated in **Table 6**.

Table 6: Relative weight of chemical parameters for WQI

Parameters	Weight (w_i)	Relative weight (Wi)	Indian Standards (mg/L; except pH) (BIS, 2012)
pH	3	0.086	6.5-8.5
TDS	2	0.057	500-2000
Fluoride	4	0.114	1.5
Chloride	3	0.086	250-1000
Nitrate	4	0.114	45
Sulphate	2	0.057	200-400
Uranium	5	0.143	0.03
Total hardness	2	0.057	200-600
Total Alkalinity	2	0.057	200-600
Arsenic	5	0.143	0.01
Iron	3	0.086	0.3
	$\sum = 35$	$\sum = 1$	

The index value ranges from 33.9 to 555. **Table 7** projects the groupings of percentages of sample in line with the water quality. Approximately 38.2 and 19.1% of groundwater samples are designated with 'poor' and 'very poor' quality, while around 9.1% of samples are termed as 'unsuitable for drinking'. Cumulatively, about 66% of water samples are not endorsed for drinking, cooking and other household activities.

Table 7. Water quality categorization based on WQI value

WQI value	Water quality	Percentage of samples
<50	Excellent	7.27
50 - 100	Good	25.5
100 - 200	Poor	38.2
200 - 300	Very Poor	19.1
>300	Unsuitable for Drinking	9.1

5.1.1.6. Human health risk assessment (HRA) induced by the four toxic elements

Health Risk Assessment study is important for quantification of future health risks mediated by ingesting the toxic components present in groundwater. Few conventional formulas are there for assessment of health risk by the United States Environmental Protection Agency or USEPA. Health risk is categorized into two classes, cancer and non-cancer which is executed through eq. (9) to (12). Non-cancer risk assessment is performed

specifically for the two non-carcinogenic noxious pollutants F^- and NO_3^- (Narsimha and Rajitha, 2018), as well as for As and U (USEPA, 2006). Risk assessment for cancerous diseases is done for As (Waqas et al., 2017). Radiological risk assessment is made for U (Ajay et al., 2016; Duggal et al., 2017). The quantification for non-carcinogenic risk is done through 'Hazard Quotient' (HQ), a proportion between 'Lifetime Average Daily Dose' (LADD) and 'Reference Dose' (R_fD). **Table 8** represents the R_fD values of chronic oral exposure dose for these four contaminants.

Table 8: Oral Reference Dose for toxic chemicals

Element	R_fD (mg/ kg/day)	Reference
As	0.0003	Rasool et al. (2015); USEPA (2005)
U	0.001	Sharma and Singh, (2016); WHO (2012)
NO_3^-	1.6	Su et al. (2013); USEPA (2001)
F^-	6×10^{-2}	Narsimha and Rajitha (2018); USEPA (1993)

The average HQ value for NO_3^- in the groundwater samples is 0.22 and the range is 0.001 to 1.29. The HQ value has crossed the acceptable level of risk in four blocks (Krishnanagar-I, Nabadwip, Kaliganj and Chapra) i.e. 1.00, 1.18, 1.20 and 1.28, respectively. The bearable level of non-cancer risk is 1 for each element (USEPA, 2005). High HQ (NO_3^-) have also been reported previously in Adimalla et al. (2019). The average HQ(F^-) is 0.10 (range: 0.005-0.42) that do not enforce any kind of health risk from F- in drinking water. Around 55% of the groundwater samples display greater HQ compared to the standard limit with an average value of 2.67. HQ value of As ranges from 0.36 to 24.6 which creates anxiety among the inhabitants that still consume As-contaminated water. Most importantly, the same population faces a serious risk from cancer for As (1.61×10^{-4} - 1.1×10^{-2}) with a mean value of 1.2×10^{-3}, way much greater than the threshold cancer risk value initiated by As, 1×10^{-6} (USEPA, 2005). This directs that the quality of drinking water has abundant potential to instigate major health problems comprising liver, lung, urinary or skin melanoma (Alam et al., 2016). According to the U concentration present in groundwater, there is no non-carcinogenic risk from U as the mean HQ is 0.14 (range: 0.008-0.75). Nonetheless the cancer risk from U exists with a mean of 2.48×10^{-3}, more than the tolerable level of radiological risk caused by U (1×10^{-4}, WHO, 2011a). A strong probability of radiation mediated adverse health hazards exists as the calculated cancer

risk range is 1.35×10^{-4} to 1×10^{-2} (**Table 9**). Sharma and Singh (2016) reported similar observations on cancer risks by radioactive U.

Table 9: Cancer and Non-Cancer Risk Assessment for the four contaminants in the study areas

Contaminants	Non-cancer risk	Cancer risk
As	(0.36 - 24.6)	(1.61×10^{-4} - 1.1×10^{-2})
F^-	(0.005 - 0.42)	-
NO_3^-	(0.001 - 1.29)	-
U	(0.008 - 0.75)	(1.35×10^{-4} - 1×10^{-2})

5.1.2. Bardhaman

5.1.2.1. Normal statisics

Considering the permissible limit of water quality parameters, the condition of Bardhaman district is shown through a normal statistical interpretation (**Table 10**).

Table 10: Normal statistics of water quality parameters in Bardhaman district

Parameter*	Unit	Min	Max	A.M.	S.D.	C.V.	Q1	Q2	Q3
pH	-----	6.25	8.15	7.47	0.4	0.05	7.28	7.54	7.76
TDS	mg/L	5	1274	325	201	0.62	187	321	442
EC	µS/cm	66	2549	700	369	0.53	439	669	895
ORP	mV	168	418	232	53.2	0.23	186	228	256
Salinity	mg/L	-	1310	296	211	0.71	160	285	410
DO	mg/L	4.55	8.36	6.33	0.8	0.12	5.80	6.21	6.74
F^-	mg/L	0.05	1.5	0.39	0.2	0.59	0.22	0.35	0.50
Cl^-	mg/L	14.06	348	64.2	54.1	0.84	32	45.68	72.9
NO_3^-	mg/L	0.4	164	8.64	24.1	2.80	0.5	0.99	4.01
SO_4^{2-}	mg/L	0.5	310	29.8	40.5	1.36	9	20.25	35.7
PO_4^{3-}	mg/L	0.05	8.98	0.42	0.7	1.79	0.1	0.27	0.5
U	µg/L	0.07	14.6	2.20	2	0.92	0.71	1.57	3.07
TH	mg/L	20	760	216	105	0.48	150	200	270
Ca^{2+}	mg/L	0.5	450	123	72.9	0.59	80	110	160
Mg^{2+}	mg/L	10	280	91.3	49.7	0.54	52.5	95	110
TA	mg/L	34.4	494	248	102	0.41	179	250	313
CO_3^{2-}	mg/L	0	138	38.4	34.8	0.91	0	34.4	62
HCO_3^-	mg/L	9.46	494	209	95.5	0.46	146	200	274
As	mg/L	<0.003	0.045	2.16	3.6	1.64	0	0	0
Fe	mg/L	0	35.1	1.64	4.1	2.48	0.1	0.33	1.43
Radiation	nSv	13	297	154	54.5	0.35	118	148.5	194

*Water quality parameters of all the samples have been analyzed based on three replicas

The pH of groundwater in the study area is between 6.25 and 8.15 with a mean value of 7.47, that indicates a tendency of water to be alkaline. Also, it is explained by the average alkalinity value 248 mg/L (range: 34.4 to 494 mg/L). Range of total hardness value is 20 to 760 mg/L with an average of 216 mg/L. The value of EC ranges between 66 and 2,549 µS/cm. The hefty variation of EC value is mostly ascribed to the geo-chemical processes predominant in thestudy area (Gupta et al., 2008). The mean TDS valueis 325 mg/L with a range of 5 to 1274 mg/L. The variation of TDS in the district is an indication of unequal groundwater tables in the region as the concentration of TDS is low with higher groundwater table and topography. Almost 87% of groundwater samples have TDS values within the permissible limit of BIS. There is a strong correlation found from regression, (R^2 = 0.71) between EC and TDS which follows general ionic rule in groundwater (**Fig. 20**). According to the classification of TDS value by Fetter (1990), ~ 2.22% of all the samples collected from the study region are of brackish nature with TDS value higher than 1000 mg/L and the remaining samples are fresh water with TDS value less than 1,000 mg/L.

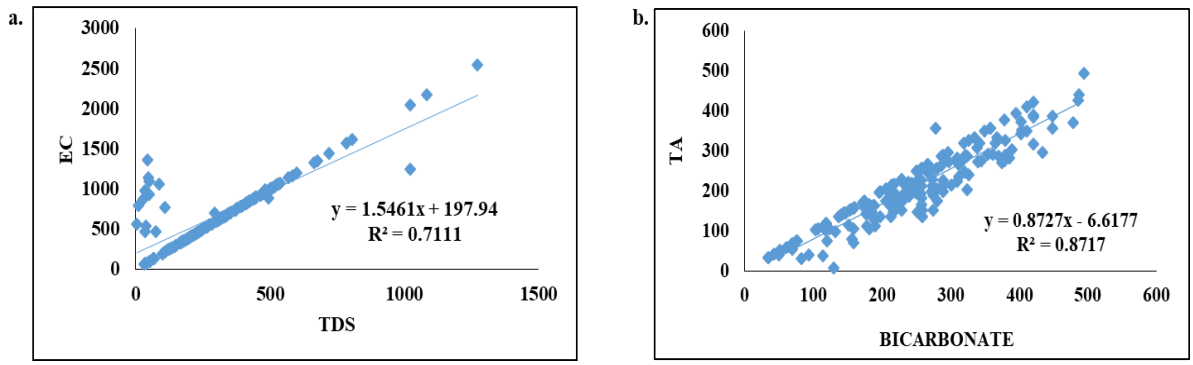


Fig. 20. Regression analysis between (a) TDS and EC, (b) Bicarbonate and Total Alkalinity in groundwater of Bardhaman

Distribution of four toxic chemical parameters (As, F^-, NO_3^- and U) in groundwater of Bardhaman district is shown in **Fig. 21**. The mean Cl-, SO_4^{2-} values are within acceptance (63.6 ± 54.2 and 32.8 ± 40.6 mg/L) butit has to be mentioned that in very few samples of some block's maximum NO_3^- concentration was observed far beyond to the acceptable limit (Fig. 21b). Maximum NO_3^- concentration was found in the samples of the blocks of West Bardhaman; Burdwan I, Ondal, Raniganj, Jamuria, Barabani and Salanpur respectively and the range was (48.4-164) mg/L. This might be a consequence of excessive use of nitrogen fertilizers in agricultural systems. It is also important to mention that the

above blocks are towards the boundary of Jharkhand which is a well-known mining area. Raniganj is itself a coal mining area.

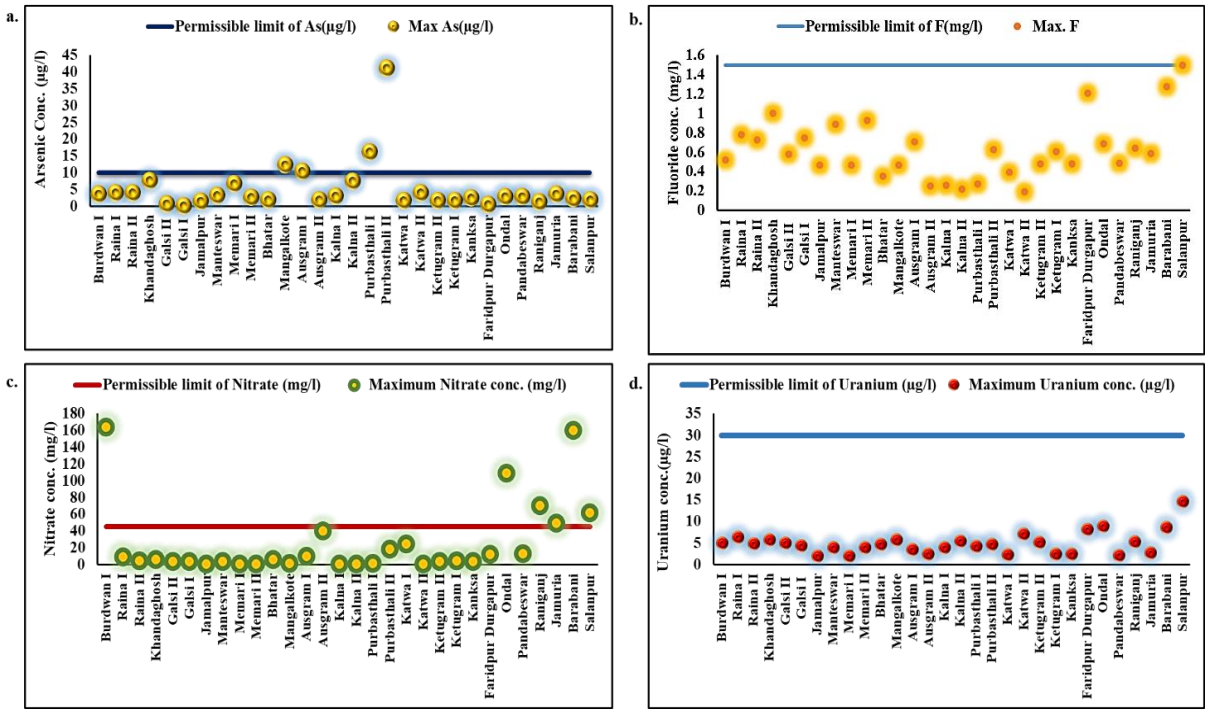


Fig. 21. Distribution of four toxic chemical parameters in groundwater in Bardhaman: a. Arsenic, b. Fluoride, c. Nitrate and d. Uranium

Another important finding is that samples from Pandabeswar block showed a maximum F- concentration of 1.5 mg/L. It might be a reason of mineral leaching from laterite soil. Total hardness increases with the increase in concentrations of calcium and magnesium ions, which is also advised by the incidence of strong positive correlations between them and also the predominance of Ca^{2+} ions over Mg^{2+} ion towards the contribution of total hardness in the study area. Total Hardness as $CaCO_3$ ranges from 20 to 760 mg/L with a mean of 216 mg/L in the study area. According to TH classification by Sawyer and Mc Carthy (1967), 58.33% samples indicate that groundwater is hard (150–300 mg/L) and 12.7% of the samples lie in the very hard (>300 mg/L) classification. 53.3% of all the collected samples Total hardness has crossed the safe limit given by BIS 10500:2012. Nearly 40% of groundwater samples own TH value above than TA value indicating the groundwater having noncarbonated hardness (Chow 1964). Such type of hardness is challenging to be eradicated from the water. Around 36. 7% of the collected samples own TH value under the acceptable limit and about 46. 7% of the collected samples have TH

under the acceptable limit as suggested by BIS. The average bicarbonate concentration and total alkalinity (TA) value is also above that of their acceptable limit in drinking water which suggests that the possible source for groundwater alkalinity comes mainly from bicarbonate ions ($R^2 = 0.87$) (Fig. 20b). It is noticed that throughout the study area [Ca^{2+}] >[Mg^{2+}] and [HCO_3^-] concentration overrules [CO_3^{2-}] concentration. If we consider only the anions present in groundwater, the range of concentration is [F^-]< [PO_4^{3-}] < [NO_3^-] < [SO_4^{2-}] < [Cl^-]. Cl^- and HCO_3^- are the dominant anions followed by Sulphate Nitrate and Phosphate. Concentrations of $HCO_3^-$and Cl^- ions are 9.46 – 494 and 14 – 347 mg/L, respectively. High concentration of Cl- ion concentration is most likely to be connected with the local anthropogenic reasons of groundwater pollution because it is associated with high NO_3^- and SO_4^{2-} contamination. It is found that most of the groundwater samples collected from shallow depth tube-wells have higher concentrations of NO^{3-}, Cl^-, and SO_4^{2-} ions. Concentration of NO_3^- in the study area ranges from 0.4 to 164 mg/L (mean 8.63 mg/L). Only 5.5% of the samples have crossed permissible limit but majority of the samples are within permissible limits. The range of iron concentration (<0.05-35.12 mg/L) proves that the groundwater of this district is iron rich. Theacceptable level of U in drinking water is 30 µg/L as per WHO (2011a) and 60 µg/L according to the Atomic Energy Regulatory Board, Department of Atomic Energy (AERB, 2004). Our study area reveals a mean U concentration of (2.18 ± 2.03 mg/L). Hence the groundwater of the present study area is safe from the adverse effect of U. In case of As, the higher concentrations are found in Purbasthali I and II block which are already reported with high As contamination in groundwater. However, the distributions of the toxic parameters in the blocks of the whole district are given in the following Table (**Table 11**).

Table 11: Block-wise statistical distribution of the four toxic chemicals in groundwater of studied area

Blocks	Sample Number (n)	Arsenic (µg/L) Range (Min-Max)	Fluoride (mg/L) Range (Min-Max)	Nitrate (mg/L) Range (Min-Max)	Uranium (µg/L) Range (Min-Max)
Ausgram I	6	0.28 - 10.36	0.05 - 0.71	0.5 - 10.16	0.07 - 3.5
Ausgram II	6	0.3 - 1.95	0.05 - 0.25	0.5 - 40	0.1 - 2.45
Barabani	6	0.96 - 2.29	0.14 - 0.49	2.86- 160	0.47 - 3.14
Bhatar	6	0.64 - 1.92	0.11 - 0.35	0.4- 5.83	0.34- 4.65
Burdwan I	10	0.46 - 3.7	0.05- 0.58	0.5 - 164	0.51 - 5.01
Faridpur Durgapur	6	0.41 - 3	0.26- 1	0.5- 11.9	0.39 - 8.16
Galsi I	6	0.01 - 0.18	0.39 - 0.75	1.7 - 3.61	2.41- 4.4
Galsi II	6	0.15- 0.43	0.37 - 0.58	1.32 - 3.56	1.2 - 5.05
Jamalpur	6	0.4- 1.66	0.17 - 0.41	0.5 - 0.66	0.33 – 2
Jamuria	6	1.26 - 3.67	0.21 - 0.6	2.29 - 49.6	0.6 - 2.81
Kalna I	6	0.64 - 2.58	0.21 - 0.26	0 - 0.5	0.65 – 4
Kalna II	6	0.7 - 7.56	0.082 - 0.22	0 - 0.5	0.35 - 2.7
Kanksa	6	1.11- 2.56	0.1 - 0.48	0.5- 3.54	0.13 - 2.46
Katwa I	6	0.64- 5.78	0.11 - 0.39	0.5 - 2.03	0.69 - 2.3
Katwa II	6	0.44 - 4.1	0.1 - 0.19	0 - 0.5	1 - 7.15
Ketugram I	6	0.86 - 1.8	0.25 - 0.61	0.5- 4.8	0.5 - 2.4
Ketugram II	6	0.62 - 1.62	0.15 - 0.48	0.5 - 3.67	0.35 - 5.15
Khandaghosh	6	3.5 - 7.69	0.31 - 1	0.8 - 5.7	1.28 - 5.76
Mangalkote	6	0.61- 12.48	0.1 - 0.47	0.5 - 1.63	0.27 - 5.72
Manteswar	6	0.53 - 3.26	0.31- 0.89	0.5 - 3.53	0.35 - 3.92
Memari I	4	0.44 - 2.27	0.29 - 0.47	0 - 0.5	0.35 - 2.05
Memari II	5	0.56 - 6.72	0.35 - 0.93	0.5 - 0.62	1.66 - 3.91
Ondal	6	0 - 3	0.35 - 0.69	1.37 - 109	1- 2.07
Pandabeswar	6	0 - 3	0.26 - 1	0.5 - 12.82	0.4 - 2.1
Purbasthali I	6	1.36 - 16.23	0.05 - 0.27	0.5 - 1.14	0.23- 4.2
Purbasthali II	6	1.14 - 41.28	0.062 - 0.63	0.5 - 18.1	0.5- 4.76
Raina I	6	3.64 - 4.14	0.31 - 0.78	0.75 - 9.24	1 - 6.38
Raina II	6	3.45 - 4	0.33 - 0.63	1.28- 4.4	2.42 - 4.94
Raniganj	6	0.7 - 2.83	0.2 - 0.64	5.28 - 70.4	0.8 - 5.28
Salanpur	6	1.13 - 2.01	0.4 -1.5	4.4- 62	0.42-14.6

5.1.2.2. Correlation matrix

The correlation matrix of all the studied WQPs is presented through a diagram (**Fig. 22**). It is noticed that pH and ORP shows a quite strong negative correlation (r = -0.47) which reveals that in the present study area, pH is negatively related to the concentration of protons/hydroxide ions in solution. It has been observed that the maximum contribution to groundwater conductance comes from chloride ions as the correlation coefficient (r) is

found to be 0.61 as well as TDS (r = 0.84). Both Ca^{2+} and Mg^{2+} contribute well to the hardness of water because in both the cases of (Ca^{2+}- TH) and (Mg^{2+}- TH) there are a significant correlation (r = 0.89 and 0.74 respectively). The involvement of SO_4^{2-} ions towards EC is understood by the mild strong correlation (r = 0.34). In this study it is important to signify that SO_4^{2-} and NO_3^- holds a good correlation (r = 0.35).

	pH	TDS	EC	ORP	Salinity	DO	F^-	Cl^-	NO_3^-	$SO4^{2-}$	PO_4^{3-}	U	TH	Ca	Mg	TA	$(CO_3)^{2-}$	$(HCO_3)^-$	As	Fe
pH	1																			
TDS	0.06	1																		
EC	0.06	**0.84**	1																	
ORP	-0.47	0.05	0.16	1																
Salinity	0.21	0.42	0.59	0.06	1															
DO	0.23	-0.05	0.00	-0.31	0.06	1														
F^-	0.26	-0.03	0.08	-0.19	-0.05	0.23	1													
Cl^-	-0.19	0.48	0.61	0.17	0.29	0.02	0.06	1												
NO_3^-	-0.36	0.16	0.24	0.13	-0.14	-0.04	0.01	0.47	1											
$SO4^{2-}$	-0.29	0.19	0.34	0.16	-0.11	-0.03	0.04	0.27	0.35	1										
PO_4^{3-}	0.09	0.09	0.04	-0.18	0.01	-0.03	0.02	-0.03	0.01	-0.07	1									
U	0.24	0.27	0.22	-0.15	0.20	0.16	0.37	0.22	0.04	0.00	-0.01	1								
TH	0.06	0.41	0.33	-0.16	-0.09	0.23	0.05	0.22	0.27	0.34	0.07	0.17	1							
Ca	0.10	0.37	0.29	-0.20	-0.09	0.26	0.10	0.21	0.26	0.28	0.05	0.20	**0.89**	1						
Mg	0.01	0.27	0.22	-0.05	-0.06	0.09	-0.07	0.15	0.17	0.24	0.09	0.08	0.74	0.37	1					
TA	0.32	0.21	0.28	-0.08	0.17	0.06	0.13	0.16	-0.05	-0.04	0.11	0.22	0.17	0.16	0.12	1				
$(CO_3)^{2-}$	0.14	0.23	0.20	-0.07	-0.12	0.09	0.21	0.16	0.09	0.00	0.03	0.29	0.38	0.39	0.24	0.40	1			
$(HCO_3)^-$	0.29	0.15	0.23	-0.05	0.23	0.03	0.05	0.11	-0.09	-0.05	0.10	0.15	0.05	0.03	0.05	**0.93**	0.07	1		
As	0.07	0.05	0.08	0.07	0.13	0.08	-0.03	-0.04	-0.05	-0.02	-0.07	-0.01	0.01	-0.02	0.03	0.06	0.04	0.05	1	
Fe	-0.25	-0.12	-0.17	-0.02	-0.11	-0.09	-0.11	-0.03	0.01	0.00	-0.06	-0.15	-0.05	-0.03	-0.05	-0.25	-0.16	-0.21	-0.02	1

Fig. 22. Correlation matrix of the physicochemical parameters in Bardhaman

5.1.2.3. Water Quality Index (WQI) assessment

WQI (eq. 1 to 4) is a reflection of the composite effect of water quality parameters which comes from the following calculation through 3 general steps. Firstly, each parameter is assigned with a weight in accordance with their relative significance in suitability of groundwater for over-all usage purposes (**Table 12**). The most toxic and radioactive elements like As and uranium are bestowed with maximum weight 5. Then the other less toxic elements like fluoride and nitrate are weighted to 4 with respect to their potentiality to cause adverse health effect.

The calculated relative weights are also given in Table 12**.**

Table 12: Relative weight calculation of chemical parameters

Parameters	Weight (wi)	Relative weight (Wi)
pH	3	0.085714286
TDS	2	0.057142857
Fluoride	4	0.114285714
Chloride	3	0.085714286
Nitrate	4	0.114285714
Sulphate	2	0.057142857
Uranium	5	0.142857143
Total hardness	2	0.057142857
Total Alkalinity	2	0.057142857
Arsenic	5	0.142857143
Iron	3	0.085714286
	∑ =35	∑ =1

The calculated WQI values are then classified into five categories of water (WQI <50, 50-100, 100-200, 200-300, >300) (**Fig. 23)** in accordance to use.

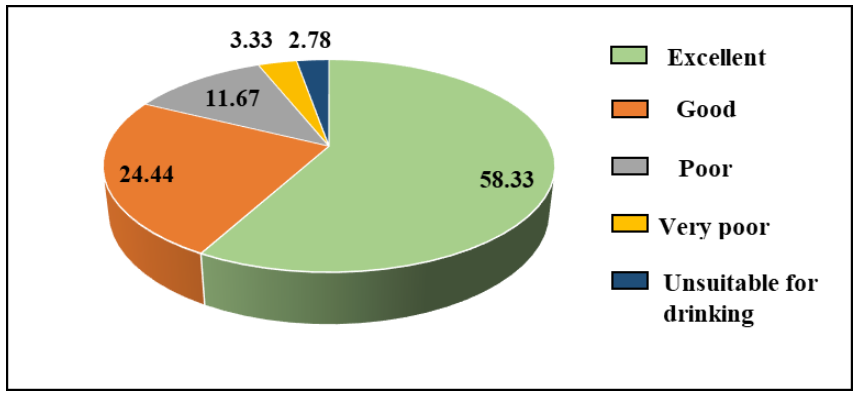


Fig. 23. WQI of groundwater samples of Bardhaman

It is well understood that the average groundwater water quality of this district is not poor. Approximately 58.3% of water can be marked as healthy for drinking and other domestic purposes. But nearly 6% of water quality is tremendously unfit for direct consumption.

5.1.3. Murshidabad

5.1.3.1. Domestic and community level shallow tube-wells

Groundwater is generally used for drinking, cooking and other domestic purposes in the study areas. Apart from domestic shallow tube-wells, groundwater is further withdrawn through government shallow tube-wells installed by the local gram panchayats in community level. The magnitude of As-contamination in drinking water throughout the study area is well demonstrated in **Table 13**. In Raninagar II, meanAs concentration value

(63.8 ± 132 µg/L) in the tube-wellwaterhaving a depth range of 30- 270 ft is approximately 6 times higher compared to the WHO recommended value in drinking water (WHO, 2011a). In Domkol block, mean As concentrations in deep and shallow tube well water are observed as 3 times higher than the WHO recommendation.

Table 13: Arsenic contamination in the tube-well withdrawn groundwater sources used for drinking and domestic purposes

Water source	Depth (ft)	Arsenic concentration (µg/L)			Iron concentration (mg/L)		
		Number of sample (n)	Mean	Range	Number of sample (n)	Mean	Range
Block: Domkol							
Deep tube well	120 - 400	65	33.4 ± 69	<3 - 422	65	1.8 ± 3	<0.01-12.8
Shallow tube well	30 - 85	50	32.5 ± 50	<3 - 231	50	1.93 ± 3.05	<0.01 – 12.2
Block: Raninagar II							
Domestic and community level shallow tube-well	30 - 270	366	63.8 ± 132	<3 - 995	360	1.60 ± 2.60	<0.05- 23.5

Mean As concentrations in tube-well water reported from two other As-exposed blocksin Murshidabad district, namely Jalangi (depth range: 20.12-64.02 m) and Domkal (depth range: 7.93-73.17 m) were 133 and 100 µg/L (range: 21-176 µg/L; n=11 and range: 6-138 µg/L; n=23), respectively (Roychowdhury et al., 2003). About 86.2% and 67.6% of hand tube-well water samples were reported with As concentration above 10 µg/L in Sagarpara (n = 565) and Raninagar-II block (n = 2211) of Murshidabad district, respectively (Rahman et al., 2005a, b). About 57% (n = 9949) and 95.6% (n = 46) of domestic tube-well water samples with As > 10 µg/L were reported from the entire Deganga block and Fakirpara village in the gram panchayet of Kolsur in Deganga block, located in North 24 Parganas district of West Bengal (Rahman et al., 2003).

Distribution of As concentrations in groundwater is evaluated with the help of a probability distribution plot (**Fig. 24a**). The study results reveal that the distribution of As is right sided tailed as most of the analyzed samples are ranged between <3-100 µg/L. The distribution is positively skewed as the values go with an order of mean (63.8 µg/L) >

median (13.9 µg/L) > mode (3 µg/L). The spatial distribution pattern of As in these tube-wells is demonstrated through contour plot based on the available sample locations or their geographical co-ordinates (**Fig. 24b**). The diagram delineates the dispersion of As in the sampling points very precisely with the help of varied color intensities (Nath et al., 2008).

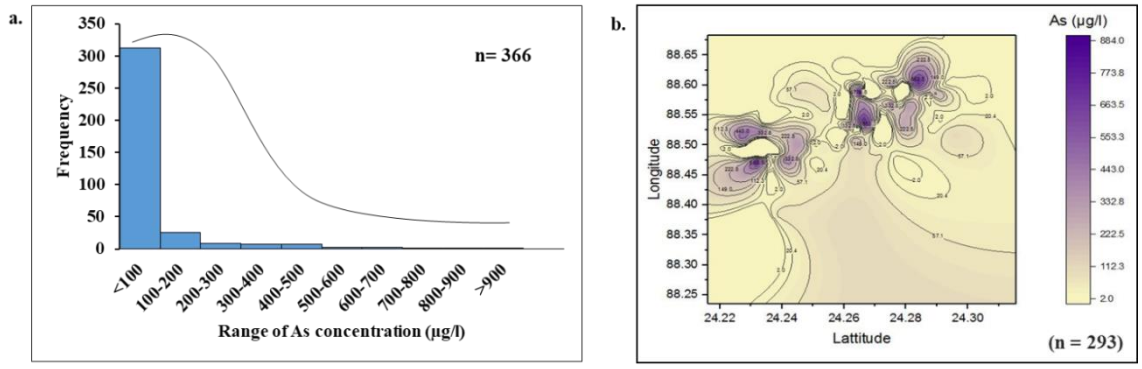


Fig. 24. Statistical distribution of arsenic in a) Domestic/community level shallow tube-wells, b) Agricultural shallow tube-wells

Distribution of As in these tube-wells from each of the 9 gram panchayat is shown in Table 2A where the percentageof higher concentrations of As is more pronounced in Malibari II gram panchayat. Approximately, 70.1, 59.4, 59, 58.9, and 56.3% of the groundwater samples exceed the permissible limit of As in drinking water in Malibari-II, Katlamari-I, Katlamari-II, Malibari-I, and Raninagar-II, respectively (**Table 14**). The groundwater As-contamination scenario in these five gram panchayats is higher compared to the other studied gram panchayats like Kalinagar-I, Kalinagar-II, Raninagar-I and Rajapur.

Table 14: Distribution of As concentration in domestic and community level shallow tube-wells

Name of the GP	No. of sample (n)	Mean As (µg/L)	Range of As (µg/L)	% of sample withAs > 10 µg/L	No. of samples with As range (µg/L)				
					≤ 10	>10-50	>50-100	>100-500	>500-1000
Malibari-I	56	56.9	<3-434	60.7	22	14	10	10	-
Malibari-II	67	141	<3-995	70.1	20	8	17	16	6
Kalinagar-I	28	39.6	3.1-387	46.4	15	5	7	1	-
Kalinagar-II	40	33.9	<3-632	42.5	23	11	4	1	1
Raninagar-I	42	18.8	<3-89.2	45.2	23	12	7	-	-
Raninagar-II	32	52.6	<3-438	56.3	14	7	8	3	-
Katlamari-I	32	87.5	<3-533	59.4	13	5	4	9	1
Katlamari-II	39	69.3	<3-833	59	16	14	2	6	1
Rajapur	30	9.08	<3-34.2	33.3	20	10	-	-	-
Total	366	63.8	<3-995	54.6	166	86	59	46	9

Arsenic concentration, its mobility and dispersion in the aquifers is more or less dependent on the depth of the aquifers (Biswas et al., 2011) and the depth itself depends on the characteristics of the aquifers along with intermediate confining clay layers (Kinniburgh et al., 2003). A detailed investigation has been done earlier regarding the enrichment of As in groundwater of the alluvial aquifers where it is stated that the variation of As with depth is caused by several factors like sediment As concentration, sorption competence of the sediment grains, the redox environment of the aquifers and volume of groundwater withdrawal (Ahmed et al., 2004). Chowdhury et al. (1999) showed that As concentration initially increases up to a certain depth (~22 m) in groundwater and then decreases with increase of the depth. Most of the agricultural tube-wells in the studied areaare installedat shallow depth zone (9.14-36.6 m) to get access of groundwater easily. The domestic and community level tube-wells available in Raninagar II have been mostly installed in shallow aquifer zone (9.14-82.3 m). In most cases, groundwater withdrawn from the shallow depth aquifer is contaminated with high concentration of As, as a result, not recommended for drinking purpose. It was perceived that the shallow depth aquifers contribute higher level of As-contamination compared to the deeper tube-wells (Chakraborti et al., 2009; Halim et al., 2010; Sengupta et al., 2004). Moreover, excessive withdrawal of water from the deeper tube-wells has eventually induced As-contamination through infiltration which makes them vulnerable too (Burgess et al., 2010; Winkel et al., 2011). This is truly corroborated in our findings, where considerably shallower depth of the agricultural aquifers has a moderately positive significance on As concentrations (R^2=0.283, Fig. 24b).

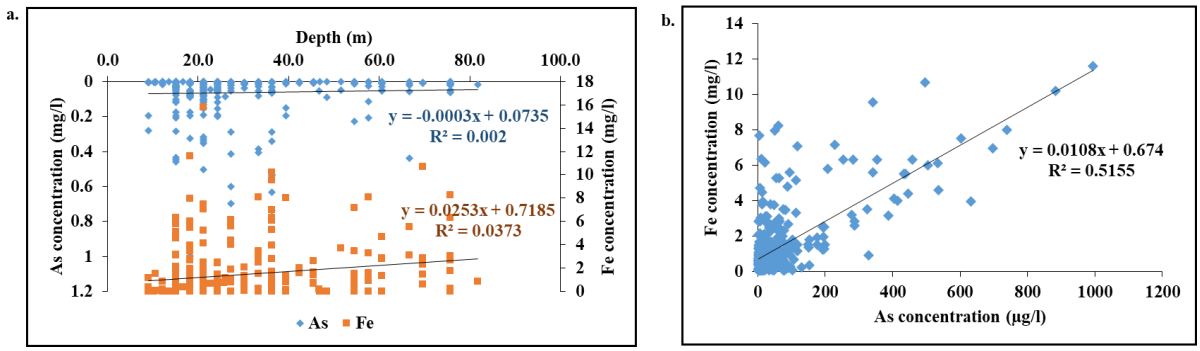


Fig. 25. Relation among depth of aquifers and groundwater As and Fe concentration: (c) Domestic and community level shallow tube-wells, (d) Agricultural shallow tube-well

Depths of the aquifers sometimes regulate groundwater Fe concentrations too. In many researches, it is observed that the well depth is inversely correlated with Fe concentration in groundwater (Duan et al., 2015; Zhang et al., 2020). Chakraborti et al. (2009) showed negative correlation between depth of the aquifers and related iron concentrationsin the groundwater samples surveyed from districts of West Bengal. Also, it is known that groundwater As is broadly attributed by underground sorption, precipitation, and dissolution processes that are controlled by redox reactions with different iron species (Root et al., 2009). It is reported that Asis likely to be co-precipitated with Fe, when water is withdrawn from underneath (Chowdhury et al., 1999). Also, Fe concentration decreases significantly when irrigational activities start in the fields i.e. groundwater withdrawal causes constant oxidation in the aquifers and the reduction rate of the iron oxides decreases accordingly (Zhang et al., 2020). Mean Fe concentration of groundwaterin the present study hasbeen observed as 1.60 mg/L (range: <0.05-23.5mg/L; n=360) in domestic and community level shallow tube-wells which is higher than the recommended value of Fe in drinking water (0.3 mg/L, BIS, 2012). Groundwater As and Fe concentrations are quite significantly inter-related to each other; the correlation valuein the domestic and community level shallow tube-wells is moderately strong (R^2=0.515) (Fig.25a). The higher mean Fe concentration in shallow agricultural aquifers and its enhanced correlation value with As clearly reveals that with continuous oxidation during enormous irrigation, Fe gets leached from the sediments and consequently infiltrates into groundwater (Anawar et al., 2003). Kar et al. (2010) also suggested that As is phenomenally associated with Fe bearing minerals as dissolved As in groundwater of Gangetic plain in West Bengal had a distinctive relationship with Fe.

5.1.3.2. Groundwater arsenic withdrawal scenario in Raninagar II

A huge amount of groundwater is exploited daily in the rural belt for household and agricultural needs beyond natural refilling of aquifers from rainfall (Dey et al., 2017). In Raninagar-II block, average number of users behind each hand tube-well was earlier observed as 14 (Rahman et al., 2005a). A large part of the studied region with an area of 206 km^2 and population approximately 1,93,118 might be at the margin of arsenical health risk through daily withdrawalof As-contaminated groundwater from 25,600 domestic and community level shallow tube-wells. The usage of groundwater for a family of four persons in the studied area is approximately 1000 L/day. Water withdrawal rate and As

withdrawn per year by the domestic and community level shallow tube-wells has been shown through the following calculations:

Water withdrawal rate of each tube-well (average) = 1000 L/day (for a family of four persons)

As withdrawn per day by each tube-well= As concentration in each tube-well× water withdrawal/day;

As withdrawn per year by each tube-well= As withdrawn per day by each tube-well× 365 days

Arsenicwithdrawal scenario from the studied areais outlined well in **Table 15**.

Table 15: Withdrawal and deposition scenario of arsenic (per year) in the 9-gram panchayats of Raninagar-II block

Name of the GP	Geographical area (km^2)	Approximate No. of domestic and community level shallow tube-wells[a]	% of sample > 10 µg/L[b]	Mean As concentration (µg/L)	As withdrawal per year through domestic tube-well> 10 µg/L (kg/year)
Malibari-I	27	3355	58.9	56.9	41
Malibari-II	17	2113	70.1	141	75.8
Kalinagar-I	21	2610	46.4	39.6	16.9
Kalinagar-II	14	1740	42.5	33.9	9.15
Raninagar-I	18	2237	45.2	18.8	6.94
Raninagar-II	16	1988	56.3	52.6	21.5
Katlamari-I	24	2982	59.4	87.5	56.6
Katlamari-II	22	2734	59	69.3	40.8
Rajapur	47	5841	33.3	9.08	6.45
Total	**206**	**25,600**	**54.6**	**-**	**275**

[a]Approximate no. of tube-wells in each GP = (total no. of tube-wells/total geographical area of the block)× geographical area of GP

[b]% of sample > 10 µg/L inanalysed samples have been extrapolated to thetotal number of tube-wells

The total As withdrawn inthe 9gram panchayatsofRaninagar-II block (∑GP) through active domestic/community purpose tube wells is 275 kg per year. The top three water withdrawing gram panchayats are Malibari II, Katlamari I and Katlamari II (75.8, 56.6 and 40.8 kg/year respectively).

5.1.3.3. Environment quality assessment of groundwater among the 9 gram panchayats

5.1.3.3.1. Single factor pollution index study for As and Fe

The results from eq. (5) show that the average As-contamination factors of the entire block for domestic and community level shallow tube-wells are high (6.62) while the Fe-contamination factor for the same are considerable (5.33). Arsenic-contamination potential of the drinking water from domestic and community level shallowtube-wells is inarguably high in Malibari-II, Katlamari-I and Katlamari-II gram panchayats with I_{As} values 14.1, 8.75 and 6.93, respectively while lowest in Rajapur (I_{As} = 0.93). Iron-contamination potential in the same water source is highest in Katlamari-I and lowest in Kalinagar-II gram panchayats with I_{Fe} values of 10.3 and 1.95, respectively.

5.1.3.3.2. Nemerow pollution index (NPI) study

Calculation from eq. (6) reveals that the groundwater quality of the study area from domestic and community level shallow tube-wells is ‘severely polluted’ (class V) as all the relevant calculated NPI values are above 3.0. The pollution index is highest in Malibari-II gram panchayat with NPI values 12.3 and 2.47, respectively. The domestic and community level shallow tube-well water from Rajapur GP attributes lowest NPI (10.0).

5.1.3.3.3. Ecological risk analysis

Unlike soil and sediments, the potential ecological risk from the aquatic systems of any site can be calculated following the method by Hakanson (1980) to estimate the future environmental crisis. Results from eq. (7) reveal that throughout the entire block, the tube-wells from the community zones or domestic levels are at high environmental risk (Er = 66.2, moderate risk). The results clearly indicate that the highest ecological risk exists in Malibari-II gram panchayat withEr values of 141 (considerable) for domestic and community based shallow tube-well. Ecological risk by domestic and community based shallow tube-wellwaterinKatlamari-I have been also observed as ‘considerable’ with an Er value of 87. 5, whereas it is ‘moderate’ in Katlamari-II (69.3), Malibari-I (57.0), Raninagar-II (52.6), and Kalinagar-I (40).

Table 16: Appraisal of environmental quality and risk for the groundwater of Raninagar II

Name of the GP	I_{As}	I_{Fe}	NPI (As + Fe)	Er (As)
Malibari-I	Considerable (5.7)	Considerable (4.32)	Severely polluted (10.6)	Moderate (57)
Malibari-II	High (14.1)	High (6.43)	Severely polluted (12.3)	Considerable (141)
Kalinagar-I	Considerable (3.96)	Considerable (4.45)	Severely polluted (10.4)	Moderate (40)
Kalinagar-II	Considerable (3.48)	Moderate (1.95)	Severely polluted (10.2)	Low (34.8)
Raninagar-I	Moderate (1.9)	Moderate (2.79)	Severely polluted (10.1)	Low (19)
Raninagar-II	Considerable (5.26)	High (7.43)	Severely polluted (10.9)	Moderate (52.6)
Katlamari-I	High (8.75)	High (10.3)	Severely polluted (12.02)	Considerable (87.5)
Katlamari-II	High (6.93)	High (8.2)	Severely polluted (11.3)	Moderate (69.3)
Rajapur	Low (0.93)	Moderate (2.38)	Severely polluted (10.0)	Low (9.29)
Total (average)	**High (6.38)**	**Considerable (5.33)**	**Severely polluted (10.8)**	**Moderate (63.8)**

5.1.3.3.4. Status of micronutrients in groundwater

Selenium (Se) and Zinc (Zn) are essential micronutrients in drinking water having anti-oxidant properties towards human health (Olivares et al., 2005; Otieno, 2017; Vinceti et al., 2013). Usually, As and Se share a mutually contrasting relationship with each other in groundwater, many a times negatively correlated (Buschman et al., 2008; Wilkin et al., 2018). Normally, in groundwater, the level of Se ranges from 0.06 µg/L to nearly 400 µg/L (Lindberg, 1968). The recommended value of Se in drinking water is set to 10 µg/L (BIS, 2012); however, in the pipeline supplied water throughout the world, Se concentration is found to be below 10 µg/L (Gore et al., 2010; WHO 2004). On the other hand, in groundwater, the normal level of Zn is 10-40 µg/L; however, in supplied tap water it may exceed the range (WHO, 2003b). The requirement level of Zn in drinking water is set as 5000 µg/L (BIS, 2012). In the present study, the mean concentrations of Se and Zn in drinking water are 0.57± 0.27 µg/L (median: 0.57 µg/L; range: 0.17-0.98 µg/L, n=12) and 84.5 ± 93.1 µg/L (median: 37.5 µg/L; range: 10-258 µg/L, n=12), respectively which state that the drinking water Se and Zn concentrations are in extreme low level compared to their recommended values. The level of both these micronutrients in groundwater from the studied area corroborates with the findings of Roychowdhury et al. (2003) where ranges of Se and Zn concentration in groundwater (n=44) were reported as <0.2-3 µg/L and <7-333 µg/L, respectively from two As exposed blocks in Murshidabad district. It is evident from our study that As plays an antithetical role with Zn in groundwater with a negative correlation (r) value (-0.30, $p > 0.05$), although, it is positively related to Se with a

correlation factor of 0.42 ($p< 0.05$). Moreover, Se and Zn are negatively dependent to each other, which is expressed through the negative correlation factor ($r= -0.33$, $p>0.05$).

Table 17: Effect of Selenium & Zinc on arsenic contamination in groundwater (correlation value; p value at 95% significance level)

r, p value	As	Se	Zn
As	1		
Se	0.419297, 0.006268	1	
Zn	-0.30409, 0.382076	-0.32755, 0.066683	1

It is important to mention here that high amount of As competes with Se in the mammalian body and inactivates Se dependent enzymes by replacing Se (Sun et al., 2014). Also, Zn is reported to increase the level ofAs detoxifying protein metallothionein (Roychowdhury, 1999; Roychowdhury et al., 2003). Thus, Se and Zn deficit in drinking water in the studied areas undeniably enhances the possibility of suffering from As toxicity and related health risks of the local inhabitants which is supported by other study (Roychowdhury et al., 2003).

5.1.3.3.5. Intake of As, Se and Zn through drinking water in the studied population

The recommended intake rate of Se varies with age, gender even pregnancy (Kuria et al., 2020). While the children between 1 to 3 years age need a minimum value of 20 µg/day, the adults and children above 14 years require 55 µg/day. Moreover, for different countries, the suggested dietary allowance range for Se is reported as (40-85) and (30-70) µg/day, respectively for adult male and females (Hurst et al., 2013) and the upper tolerable limit is recommended as 400 µg/day (FAO/WHO, 1998; NAS, 2000). It is important to discuss that Se supplementation is known in association to reduction of As induced toxicity in organisms (Sun et al., 2014) and lowering of cancer risk (Combs et al., 1997). In Southeast Asian countries, where Se deficiency is prevalent, symptoms of arsenicosis in humans are more prominent (Roychowdhury et al., 2003; Smith et al., 2000). However, previous findings also state that excessive supplementation of Se escalates the risk of some detrimental diseases like dermatitis, non-melanoma skin cancer, alopecia, prostate cancer, type-2 diabetes etc. (Rayman, 20012, 2017). Zn is also a nutritional component for human health (Plum et al., 2010) and it too is reported with As toxicity reduction potency (Modi

et al., 2005). According to Joint FAO/WHO Expert Committee on Food Additives (JECFA), the provisional maximum tolerable daily intake (PMTDI) value of Zn is 1.0 mg/kg of body weight and the recommended daily requirement is 0.3 mg/kg of body weight (JECFA, 1982). Our study resultsreveal that the daily intake of As through drinking water among the populations in Raninagar-II block is much higher compared to Se and Zn, however, Se being higher than Zn (**Table 18**). Since, Se and Zn deficiency promotes As toxicity, an adult male individual in this study might have increased health risk from an intake of 5.52 µg/kg bw/day of As along with poor Se and Zn intake rate of 0.048 and 0.007 µg/kg bw/day, respectively. Similarly, the trend of daily intake rate for an adult female in this studyis 4.81, 0.041 and 0.006 µg/kg bw/day for As, Se and Zn, respectively. Recommended daily intake rate of Se has been set at 0.9 mg/kg of body wt. for adults while that for Zn is 15 mg/day (adult male) and 12 mg/day (adult female) (NAS, 1989; Rubio et al., 2009). The Provisional Tolerable Daily Intake (PTDI) value of inorganic As is known to be 3 µg/kg bw/day (WHO, 2011b). In addition to drinking water, highly As-contaminated rice grains are evidenced to have reduced amount of essential micro-nutrients like Se, Zn and Ni which consequences into poor quality of diet (Chowdhury et al., 2020a; Roychowdhury et al., 2003; Williams et al., 2009).

Table 18: Intake of As, Se and Zn (per day) through drinking water

Group	Daily water consumption (L/day)	Mean concentration in domestic and community level shallow tube-well			Daily intake (µg/day)			Intakerate (µg/kg bw/day)		
		As (µg/L)	Se (µg/L)	Zn (mg/L)	As	Se	Zn	As	Se	Zn
Adult male	5	63.8	0.57	0.084	331	2.85	0.42	5.52	0.048	0.007
Adult female	4	63.8	0.57	0.084	265	2.28	0.34	4.81	0.041	0.006

5.1.3.3.6. Health risk assessment

5.1.3.3.6.1. Risk assessment through consumption of drinking water from domestic and community level shallow tube-wells in all the 9 gram panchayats

The health risk assessment study according to USEPA model (eq. 9 to 11) shows that the population of the study area faces serious health threat due to prolonged exposure of As-contaminated drinking water. Continual oral intake along with nominal dermal exposure causes lifetime cancer risk in the inhabitants. The range of $ADD_{ingestion}$ values for adult

males and females in the entire block are 0.774-11.75 and 0.676-10.26 mg/kg bw/day, while the range of ADD_{dermal} values are 1.39×10^{-3} - 2.115×10^{-2} ,and 1.52×10^{-3} - 2.3×10^{-2} mg/kg bw/day, respectively (**Table 19**). The population from each gram panchayat havea clear risk of cancer as the TCR values are way much higher than the usual acceptable range of risk (1×10^{-6} to 1×10^{-4}) (USEPA, 2005) and importantly, the risk is highest in studied adult males than adult females (**Fig. 26**). Total cancer risk is found least in Rajapur (1.17×10^{-3} for male and 1.02×10^{-3} for female) and maximum in Malibari-II gram panchayat (1.77×10^{-2} for male and 1.54×10^{-2} for female). In continuation with the consumption of poor quality of drinking water, the average TCR value for adult males and females in the entire block is 8.01×10^{-3} and 7.00×10^{-3}, much higher than thethreshold limit(1×10^{-6}, USEPA, 2005). Proportion of cancer risk from dermal exposure is negligible in comparison to the oral exposure; the average CR_{dermal} values in the entire block for adult male and adult female are 3.50×10^{-5} and 3.82×10^{-5}, whereas, $CR_{ingestion}$ values for studied adult male and adult female are 7.97×10^{-3} and 6.96×10^{-3}, respectively. On the other hand, possibilities of severalnon-carcinogenichealth issues existin these studied adult individuals as the respective HI values are beyond allowance i.e. 1in each of the gram panchayat. Average $HQ_{ingestion}$ and HQ_{dermal}values for adult males and females in the entire block are 17.72, 15.46, and0.050, 0.054, respectively. The estimated ranges of non-cancer risk or hazard index value (HI) in adult males and females in this block are (2.58-39.28) and (2.26-34.3), respectively and the risk of non-cancer is highest for studied adult males than females. The average HI value in the block is shockingly 17.8 and 15.5 times higher than thethreshold value in adult males and females. Arsenic toxicity induced health risk in the study area is highest in Malibari-II followed by Katlamari-I, Katlamari-II, Malibari-I, Raninagar-II, Kalinagar-I, Kalinagar-II, Raninagar-I and Rajapur.

Table 19: Cancer and non-cancer risk evaluation in the study area with respect to As-contaminated drinking water from domestic and community level tube-wells

Name of the GP	Male			Female			Male			Female		
	$CR_{ingestion}$	CR_{dermal}	TCR	$CR_{ingestion}$	CR_{dermal}	TCR	$HQ_{ingestion}$	HQ_{dermal}	HI	$HQ_{ingestion}$	HQ_{dermal}	HI
Malibari I	7.12×10^{-3}	3.13×10^{-5}	7.16×10^{-3}	6.22×10^{-3}	3.41×10^{-5}	6.25×10^{-3}	15.83	0.045	15.87	13.81	0.049	13.86
Malibari-II	1.76×10^{-2}	7.74×10^{-5}	1.77×10^{-2}	1.53×10^{-2}	8.44×10^{-5}	1.54×10^{-2}	39.17	0.11	39.28	34.18	0.121	34.30
Kalinagar- I	4.95×10^{-3}	2.17×10^{-5}	4.97×10^{-3}	4.32×10^{-3}	2.37×10^{-5}	4.34×10^{-3}	11.00	0.031	11.03	9.6	0.034	9.63
Kalinagar- II	4.35×10^{-3}	1.91×10^{-5}	4.7×10^{-3}	3.8×10^{-3}	2.08×10^{-5}	3.82×10^{-3}	9.67	0.027	9.69	8.43	0.029	8.46
Raninagar I	2.37×10^{-3}	1.04×10^{-5}	2.39×10^{-3}	2.07×10^{-3}	1.14×10^{-5}	2.08×10^{-3}	5.28	0.015	5.29	4.60	0.016	4.62
Raninagar-II	6.58×10^{-3}	2.89×10^{-5}	6.62×10^{-3}	5.75×10^{-3}	3.16×10^{-5}	5.78×10^{-3}	14.64	0.041	14.68	12.77	0.045	12.82
Katlamari- I	1.09×10^{-2}	4.81×10^{-5}	1.10×10^{-2}	9.57×10^{-3}	5.25×10^{-5}	9.62×10^{-3}	24.36	0.069	24.43	21.26	0.075	21.33
Katlamari-II	8.62×10^{-3}	3.79×10^{-5}	8.66×10^{-3}	7.53×10^{-3}	4.13×10^{-5}	7.57×10^{-3}	19.17	0.054	19.22	16.72	0.059	16.78
Rajapur	1.16×10^{-3}	5.1×10^{-6}	1.17×10^{-3}	1.01×10^{-3}	5.56×10^{-6}	1.02×10^{-3}	2.58	0.007	2.58	2.25	0.008	2.26
Total (average)	7.97×10^{-3}	3.50×10^{-5}	8.01×10^{-3}	6.96×10^{-3}	3.82×10^{-5}	7.00×10^{-3}	17.72	0.050	17.77	15.46	0.054	15.52

5.2. Groundwater quality evaluation for irrigation purposes

5.2.1. Irrigational water arsenic contamination

The shallow tube-wells of depth range 9.14 to 36.6 mused for irrigation purposes in Raninagar II, Murshidabad, showed a mean As concentration of 138 ± 186 µg/L (n=67), certainly bigger than the commended value of As in irrigational wateri.e. 100 µg/L (CCME, 1999; FAO, 1985; Jones et al., 2017). The distribution of As concentrations in groundwater of the shallow depth tube-wells is shown through a spatial distribution plot along with a histogram (**Fig. 26**).

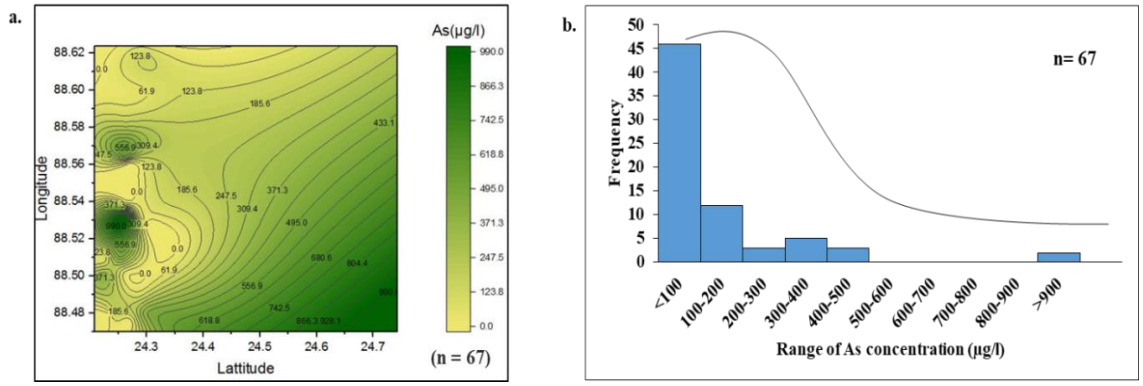


Fig. 26. Arsenic contamination in irrigational water in Raninagar II, Murshidabad: (a) Spatial distribution, (b) Statistical distribution

The range of As concentration in these aquifers (<3-990 µg/L) is articulated through the spatial distribution plot. The condensed contour lines indicatehigher concentrations with intensified colors. The maximum Asconcentration in irrigational water is observed at Malibari II gram panchayat (Lat-24° 15' 54.072" N; Long-88° 31' 58.044" E). It is wellunderstood that over the aquifers, As concentrations are clustered towards comparatively lower range (<100-500 µg/L) and thus the distribution is positively skewed or right sided tailed. Highest frequency is observed at <100 µg/L followed by (100-200) and (300-400) µg/L, respectively. Higher As concentration was reported in irrigationa lshallow tube-wells from Jalangi (range: 40-182 µg/L; n= 4) and Domkal block (range: 18-200 µg/L; n=6) of Murshidabad district, respectively (Roychowdhury et al., 2003). Distribution of As in these tube-wells from each of the 9 gram panchayat is shown in **Table 20**; As contamination scenario is maximum in Raninagar-II (80%), followed by Malibari-II (62.5%) and Malibari-I (42.9%) which exceeds the recommended limit in irrigational water. Apart from the indigenous As concentration, the tertiary provenance of As in groundwater is the irrigational return flow from the agricultural fields (Chakraborty et al., 2015).

Table 20: Distribution of arsenic in agricultural shallow tube-wells

Name of the GP	No. of sample (n)	Mean As (µg/L)	Range of As (µg/L)	% of sample with As > 100 µg/L	No. of samples with As range (µg/L)		
					< 100	>100-500	>500-1000
Malibari-I	14	130	3.52-371	42.9	8	6	-
Malibari-II	16	294	69.7-990	62.5	6	8	2
Kalinagar-I	7	27.1	11.2-46.3	-	7	-	-
Kalinagar-II	5	47.9	33.3-63.2	-	5	-	-
Raninagar-I	1	25.4	25.4	-	1	-	-
Raninagar-II	5	212	75.1-410	80	1	4	-
Katlamari-I	7	89.5	3.36-303	28.6	5	2	-
Katlamari-II	6	47	<3-122	33.3	4	2	-
Rajapur	6	53	3.2-186	16.7	5	1	-
Total	67	138	<3 - 990	37.3	42	23	2

It is known that groundwater As is broadly attributed by underground sorption, precipitation, and dissolution processes that are controlled by redox reactions with different iron species (Root et al., 2009). It is reported that As is likely to be co-precipitated with iron, when water is withdrawn from underneath (Chowdhury et al., 1999). Also, iron concentration drops significantly when irrigational activities start in the fields i.e. groundwater withdrawal causes constant oxidation in the aquifers and the reduction rate of the iron oxides decreases accordingly (Zhang et al., 2020). Mean Fe concentration for agricultural shallow tube-wells is 2.77 mg/L (range: <0.05-15.5mg/L; n=66) which is however within the recommended limit of Fe in irrigational water, 5.0 mg/L (Fipps 2003; Sharifi and Sinegani, 2012). Groundwater As and Fe concentrations are quite significantly inter-related to each other; the correlation is stronger in thecase of agricultural shallowtube-wells (R^2=0.816) **(Fig. 27).** The higher mean Fe concentration in shallow agricultural aquifers and its enhanced correlation value with As clearly reveals that with continuous oxidation during enormous irrigation, Fe gets leached from the sediments and consequently infiltrates into groundwater (Anawar et al., 2003). Kar et al. (2010) also suggested that As is phenomenally associated with Fe bearing minerals as dissolved As in groundwater of Gangetic plain in West Bengal had a distinctive relationship with Fe.

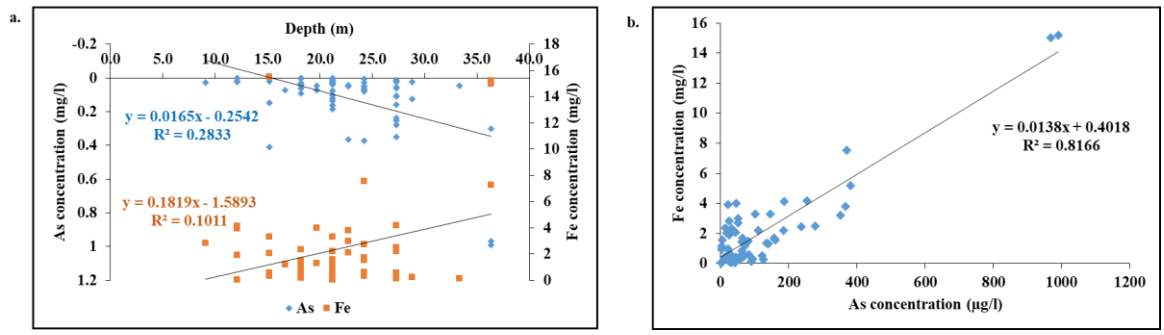


Fig. 27. Relation among depth of the tube-wells, arsenic and iron concentrations

5.2.2. Groundwater arsenic withdrawal scenario in the study area

A huge amount of groundwater is exploited daily in the rural belt for household and agricultural needs beyond natural refilling of aquifers from rainfall (Dey et al., 2017). In Raninagar-II block, average number of users behind each hand tube-well was earlier observed as 14 (Rahman et al., 2005a). A large population approximately 1,93,118 might be at the margin of arsenical health risk through daily withdrawal of As-contaminated groundwater from 10,100 agricultural shallowtube-wells (Assistant Director of Agriculture, 2019). The average discharge rate of each shallow tube-well in an agricultural field is 20 m^3/h which runs approximately for 7h/day and 7months/year, especially during the pre-monsoon cultivation. Water withdrawal rate and As withdrawn per year by the agricultural shallow tube-wells have been shown through the following calculations:

Water discharge rate of each tube-well (average) = 20 m^3/h

Daily water discharge rate of each tube-well (average) = 20 m^3/h× 7h/day = 140 m^3/day

Yearly water discharge rate of each tube-well (average) = 140 m^3/day × 210 days (7months/year)

= 140×210 m^3/year = 140×210×1000 L/year

= 2.94×10^7 L/year (Roychowdhury et al., 2005)

Arsenic withdrawn per year by each tube-well = arsenic concentration in each tube-well × water discharge rate/year

(% of both kind of sample with As concentration above their respective recommended limits have been extrapolated to their total number of tube-wells)

Arsenic withdrawal scenario from the studied area is described well in **Table 21**.

Table 21: Yearly arsenic withdrawal and deposition scenario in the 9 gram panchayats of Raninagar-II block

Name of the GP	Geographical area (km^2)	Irrigated area (km^2)	Approximate No. of agricultural shallow tube-wells [a]	% of sample > 100 µg/L [b]	Mean As concentration (µg/L)	As withdrawal per year through agricultural shallow tube-well> 100 µg/L (kg/year)	As deposited on the irrigational field per year (kg/ha)
Malibari-I	27	15.2	1324	42.9	130	2171	1.43
Malibari-II	17	9.57	833	62.5	294	4500	4.70
Kalinagar-I	21	11.82	1030	-	27.1	-	-
Kalinagar-II	14	7.88	686	-	47.9	-	-
Raninagar-I	18	10.14	883	-	25.4	-	-
Raninagar-II	16	9.01	784	80	212	3909	4.34
Katlamari-I	24	13.51	1177	28.6	89.5	886	0.66
Katlamari-II	22	12.4	1079	33.3	47	496	0.40
Rajapur	47	26.46	2304	16.7	53	599	0.23
Total	**206**	**116**	**10,100**	**37.3**	**-**	**12561 = 12.5 tonnes**	**1.08**

[a]Approximate no. of tube-wells in each GP = (total no. of tube-wells/total geographical area of the block) × geographical area of GP

[b]% of sample > 100µg/Linanalysed samples have been extrapolated to the total number of tube-wells

In this study, As withdrawal through shallow tube-wells was calculated based on the recommended value of As in irrigational water as 100 µg/L (CCME, 1999). The As withdrawn from the shallow tube-wells of the agricultural fields of the 9 gram panchayats of Raninagar-II block (∑GP) is 12.5 tonnes. Arsenic withdrawal in three blocks namely Kalinagar-I, Kalinagar-II and Raninagar-I through irrigational water have not been considered as no sample was observed with As concentration above permissible limit. Roychowdhury et al. (2005) reported that approximately 1.85 and 0.56 kg/year of As were withdrawn with groundwater from two respective shallow tube-wells used for irrigational purposes in As-affected villages of Raninagar-II block (Rakhaldaspur and Dobopara, respectively). Arsenic withdrawn by the 19 shallow tube-wells was reported in the range of 0.53-5.88 kg/year from Domkal block of Murshidabad (Roychowdhury et al., 2002). Mandal (1998) reported that about 0.176 and 6.4 tonnes of As were withdrawn per yearfrom all the domestic hand tube-wells (n=9190) and agricultural shallow tube-wells (n=3072), respectively from Deganga, another severely As-effected block located in North 24 Parganas district of West Bengal. Hence, a substantial amount of As is accumulated in

the food chain through the irrigated water. All these findings support our study results and explain that As withdrawal scenario varies drastically in different districts and also increases with increasing groundwater withdrawal rate and time. According to Mandal (1998), approximate 6.4 tonnes of As per year was withdrawn from a single block considering the recommended limit of As in irrigational water as 10 µg/L. Following the same trend, the withdrawal of As in the entire block of this study would be equal toas much as 37.4 tonnes/year.

5.2.3. Arsenic deposition on the agricultural fields

The total irrigated area used for agriculture in Raninagar-II block is 11600 ha per year (Assistant Director of agriculture, 2019). Average Asdeposition rate in the agricultural fields of the studied areais estimated to be 1.08 kg/ha (range: 0.23-4.70 kg/ha). The results are confirmed using a correlation matrix among the variables. It depicts that deposition of Ason soil is majorly correlated with the As concentration in irrigational water ($r = 0.97$). Moreover, As concentrations in these aquifers also have a significant contribution behind the annual As withdrawn in the studied area ($r = 0.99$). Roychowdhury (2008) showed deposition of As in the agricultural lands ($n= 23$) of different affected blocks of Murshidabad district in therange of 1.66-16.8 kg/ha considering recommended As concentration in groundwater as 10 µg/L. According to a study by Meharg and Rahman (2003) in Bangladesh, the withdrawn As from aquifers retains in the first 10 cm of soil when the irrigation water contains 100 µg/L of As (assuming soil density of 1 kg/L), consequently the water input would cause a yearly increase in soil As by 1 mg/kg.In another study in Bangladesh, it is reported that about 71% of the total As that is added to the rice field comes from the As accumulated in the top 0-75 mm layer of soil which is accounted by irrigation water (Saha and Ali, 2007). Therefore, food chain As-contamination is a burning issue as the huge amount of Aswithdrawal gets deposited in soil sediment and an ample portion of it gets accumulated in plant rhizosphere and thus translocated into crops and grains (Chowdhury et al., 2018a, 2020b, Roychowdhury et al., 2005). Many researches proved that irrigational water increases indigenous soil As concentration during dry season (Chowdhury et al., 2018a; Shrivastava et al., 2014; Ullah,

1998); however, after the monsoonal cultivation season, when the paddy fields are inundated with rain water, the surface soil-As concentrations decreases extensively (Chowdhury et al., 2020b; Saha and Ali, 2007).

5.2.4. Effect on paddy and rice grain

Murshidabad is an agriculture dependent district with 204.3 ha of net irrigated area and 398.7 ha of net sown area where the cropping intensity is reported as 245% (http://agricoop.nic.in/sites/default/files/WestBengal%2011-Murshidabad-31.12.2011.pdf). Paddy, wheat, maize, jute, potato and lentils are the most cultivated crops and vegetables in the district and the fields are frequently irrigated with groundwater. Considerable amounts of As was reported in the rice grain (mean: 239 µg/kg; range: 43-662 µg/kg; n=34) cultivated using As-contaminated groundwater (mean: 75 µg/L; range: 18-200 µg/L; n=23) in Jalangi, Domkal, Hariharpara and Raninagar-II blocks of Murshidabad district (Roychowdhury, 2008). Higher As accumulation was reported in different parts of paddy plant with increasing doses, irrespective of the cultivar (Bhattacharya et al., 2013). The estimated mean As concentrationsin paddy whole grains and rice grains from the studied area are 369 ± 222 µg/kg (range: 124-800 µg/kg; n=25) and 247± 183 µg/kg (range: 52-668 µg/kg; n=25), respectively. A broad range of As concentration (<0.3-2600 µg/kg) was observed in the rice grain among 44 cultivars in Nadia district of West Bengal (Samal et al., 2021). Samal et al. (2011) reported the presence of considerable amounts of As in rice grains cultivated from two different seasons, Aman (mean = 194 µg/kg; n = 19) and Boro (mean = 156 µg/kg; n = 16) in the same district. Assuming an average annual rice production rate of 2678 kg/ha in the Murshidabad district (http://agricoop.nic.in/sites/default/files/WestBengal%2011-Murshidabad-31.12.2011.pdf), total As accumulationin rice cultivated perland per year is 1.98 g/ha and that of paddy whole grain is 4.24 g/ha (**Table 22**).

Table 22: The effect of deposited arsenic on agricultural lands on rice and paddy grain

Parameter	Quantity
Productivity of rice per land (kg/ha)[a]	
Rabi	2024
Kharif	2510
Summer	3478
Average	2678
Effect of As contaminated irrigational water in rice grain	
As concentration in rice grain	247 µg/kg
Total As accumulation in rice grain per land per year (in three different cultivation seasons)	**(**2678 × 247× 3) = 1.98 g/ha
As deposition per land per year	1.08 kg/ha
Accumulation of As in rice grain from deposited land	(1.98 g/ha)/(1.08 kg/ha)× 100 % = 0.18%
Effect of As contaminated irrigational water in whole paddy grain	
Weight ratio for production of rice grain from paddy grain[b]	28 kg rice from 40 kg paddy
Productivity of paddy grain per land (kg/ha)	(2678 ×40/28) = 3826
As concentration in paddy grain	369 µg/kg
Total As accumulation in paddy whole grain per land per year (in three different cultivation seasons)	(3826 ×369× 3) = 4.24g/ha
As deposition per land per year	1.08 kg/ha
Accumulation of As in paddy grain from deposited land	(4.24 g/ha)/(1.08 kg/ha) × 100 = 0.39%
Bioaccumulation factor	
Rice grain	1.78
Whole paddy grain	2.67

[a]*http://agricoop.nic.in/sites/default/files/WestBengal%2011-Murshidabad-31.12.2011.pdf*

[b]Roychowdhury (2008)

Arsenic accumulation percentage in whole paddy grain (0.39%) from deposited land through groundwater is calculated to be more than twice than that of rice grain (0.18%). The direct effect of contaminated irrigational groundwater in cultivated crops is computed through bioaccumulation factor or Bio accumulation factor (BAF).

5.2.4.1. Bioaccumulation of arsenic in paddy and rice grain

Bioaccumulation factor (BAF) is estimated to understand the extent of assimilation of any toxic environmental component in plant materials or foodstuffs from contaminated soil or

water (Chowdhury et al., 2020b; Yadav et al., 2017). The calculation is done for rice and paddy grains with respect to groundwater following:

$BAF = C_{rg}$ or C_{pg}/C_{igw} eq. (16)

Where, C_{rg}, C_{pg} and C_{igw} are denoted by concentration of As in rice grain, paddy grain and irrigational groundwater, respectively.

The BAF of whole paddy grains is also observed to be higher (2.67) than rice grains (1.78) in the study area which explains their potential of hyper-accumulation (BAF ≥ 1) (Samal et al., 2021). The higher Asaccumulation in whole paddy grain than rice grain is a clear consequence of greater extent of bioaccumulation in paddy husk, which is supported by our previous study (Chowdhury et al., 2018b). The assimilation of high As in paddy husk causes a significant health risk in domestic livestock through their daily diets which indirectly enhances the risk of human health and environment.

5.2.5. Environment quality assessment of irrigational water among the 9 gram panchayats

5.2.5.1. Single factor pollution index study for As and Fe

A contamination assessment has been done applying thesingle factor pollution index scheme forthe water quality of the 9 gram panchayats based onAs and Fe concentrations in groundwater along with a classification of pollution in an organized way (Hakanson, 1980). A single factor pollution index (Ii) assessment has been done to evaluate the groundwater contamination with special reference to As and Fe following the equation (Zhang et al., 2018).

The results show that the average As-contamination factors of the entire block for agricultural shallow tube-wells is moderate (1.33) while the Fe-contamination factors for the same are low (0.55). In case of agricultural shallowtube-well water, the degree of contamination potential is comparatively lesser than the former one (**Table 23**). The I_{As} in Malibari-II, Raninagar-II and Malibari-I gram panchayat (2.94, 2.12 and 1.35) and the moderate I_{Fe} values are observed in Raninagar-I and Raninagar-II (1.35 and 1.16), respectively.

5.2.5.2. Nemerow pollution index (NPI) study

It is a comprehensive pollution index study based on the single factor pollution index. It is used to assess the water quality of different sampling sites which simultaneously highlights

the importance of several metals in groundwater (Bodrud-Doza et al., 2019; Nemerow, 1974; Zhong et al., 2015). Nemerow pollution indexing in groundwater of the studied area is performed considering As and Fe concentrations in groundwater. Irrigational water in each of the 9-gram panchayat is estimated as 'heavily polluted' (class IV) as per the critical limits of NPI ($2.0 \leq NPI < 3.0$) (Yan et al., 2015). The NPI is highest in Malibari-II gram panchayat with 2.47 (heavily polluted) and lowest in Kalinagar-I and II (2.09, heavily polluted) (Table 23).

5.2.5.3. Ecological risk analysis

Unlike soil and sediments, the potential ecological risk from the aquatic systems of any site can be calculated following the method by Hakanson (1980) to estimate the future environmental crisis. The potential ecological risk assessment evaluates the potential impact of metals on ecosystems (Egbueri, 2020a, b). Ecological risk is dependent on heavy metal concentrations in any system, type of contaminant and strength of toxicity. The results reveal that throughout the entire block, the tube-wells from agricultural fields produce low risk comparatively (Er = 13.3). The results clearly indicate that the highestecological risk exists inMalibari-II gram panchayat withEr value of 29.4 for agricultural shallow tube-well.

Table 23: Assessment of environmental quality and risk for the groundwater of Raninagar II block

Name of the GP	I_{As}	I_{Fe}	NPI (As + Fe)	Er (As)
Malibari-I	Moderate (1.3)	Low (0.42)	Heavily polluted (2.17)	Low (13)
Malibari-II	Moderate (2.94)	Low (0.82)	Heavily polluted (2.47)	Low (29.4)
Kalinagar-I	Low (0.27)	Low (0.33)	Heavily polluted (2.09)	Low (2.71)
Kalinagar-II	Low (0.47)	Low (0.072)	Heavily polluted (2.09)	Low (4.66)
Raninagar-I	Low (0.25)	Moderate (1.35)	Heavily polluted (2.16)	Low (2.54)
Raninagar-II	Moderate (2.12)	Moderate (1.16)	Heavily polluted (2.38)	Low (21.2)
Katlamari-I	Low (0.90)	Low (0.45)	Heavily polluted (2.13)	Low (8.95)
Katlamari-II	Low (0.47)	Low (0.28)	Heavily polluted (2.12)	Low (4.7)
Rajapur	Low (0.49)	Low (0.38)	Heavily polluted (2.10)	Low (4.85)
Total (average)	**Moderate (1.38)**	**Low (0.55)**	**Heavily polluted (2.19)**	**Low (13.8)**

5.3. Food chain arsenic contamination with special reference to rice

5.3.1. Height wise arsenic accretion and translocation in different fragments of whole paddy grain in a single pedicel

Throughout the Bengal delta, rice is cultivated in two seasons generally; Boro cultivation in pre-monsoon with natural groundwater and Aman cultivation in monsoon using groundwater due to inadequate rainfall. Around 5.8 million ha in West Bengal is under rice cultivation that covers both irrigated and the rain-fed areas, with 2.6 tonnes/ha average productivity. Most of the area under rice cultivation in Bengal is As-affected resulting into the entry of As in plants grown in contaminated soil and irrigational water (Chowdhury et al., 2018). Liu et al. (2006) showed that As reasons phyto-toxicity in paddy plants by accumulation in root, shoot, grain through translocation from the rhizosphere. Williams et al. (2007) stated that arsenic is absorbed by rice by ~ 10 times more than the other cereal crops. The typical translocation factor for As is around 0.8, which is more than barley and wheat (0.2 and 0.1) (Kalita et al., 2018). Many studies (Abedin et al., 2002; Roychowdhury, 2008) established that As translocation follows a definite pattern in paddy plant system, i.e. root>stem>leaf >pedicel>grain; however, the exact purpose is still a subject of interest. As per Zhao et al. (2012), the law of translocation has been recognized with use of radioactive As^{73} tracer where it is established that near 3% of As is accrued in grains from roots. It is noticed throughout our previous researches that As translocates in grains from roots, with concentrations declining by quite a few orders in both the As contaminated and non-contaminated area (Chowdhury et al., 2018a, 2020b). The current study examines the accretion and translocation of As in rice grains in a single pedicel of paddy plants that has been executed in both As exposed and unexposed sites (**Table 24**).

Table 24: Height wise arsenic concentration (µg/kg) in different parts of whole paddy grain located in a single pedicel

		Whole paddy grain			Rice grain			Rice husk		
		Top	**Mid**	**Low**	**Top**	**Mid**	**Low**	**Top**	**Mid**	**Low**
Exposed site Teghoria	**Mean**	246	283	307	220	257	272	578	721	888
	SD	50.4	108	104	63.2	101	72.6	121.3	236	471
	Range	207-332	206-457	190-446	172-318	169-407	195-353	385-682	413-1046	469-1642
Exposed site Madhusudankati	**Mean**	195	198	230	162	176	205	425	514	570
	SD	13.8	18.8	26.4	20.4	29.2	33.5	46.2	93.5	123
	Range	181-211	169-214	213-276	130-184	133-209	160-244	366-479	377-615	470-779
Control site Pingla	**Mean**	137	116	176	156	115	162	250	290	391
	SD	54.5	19.0	63.6	74.6	31.6	52.1	82.9	56.5	87.5
	Range	90.5-230	93-123	98.6-241	106-286	84.6-163	104-205	212-413	202-350	301-508

Height wise, As accretion follows the particular trend from lower grains to the upper grains in As exposed sites and control site (**Fig. 28a**). As concentration in rice grain is higher in the fields of Teghoria (range: 169-407 µg/kg; mean: 249 µg/kg) than Madhusudankati (range: 130-244 µg/kg; mean: 181 µg/kg). The average As concentration is found to be maximum in the rice grains of lower portion of the pedicels followed by middle and top part (272, 205 and 162 µg/kg; 257, 176and 156 µg/kg; 220, 162 and 115 µg/kg) respectively in Teghoria, Madhusudankathi and Pingla fields (Fig. 28a). The concept of translocation is also sustained in the present study for paddy husk and whole grains where husk hold higher As compared to whole paddy and rice grains. In the lower portion of the pedicel, average As concentrations of whole grain and rice husk in the three fields are 307 and 888 µg/kg, 230 and 570 µg/kg, 176 and 390 µg/kg, respectively.

The reduction % of As in paddy from lower portion of the pedicel to the upper portion is found to be less in the As exposed fields than control sites (**Fig. 28b**). In As-exposed paddy fields, the average As decrease % in whole paddy grains from low to top (of a single pedicel) is found to be 17.6, lesser than the control site fields (34%). The decay % of As from the lower pedicel to top in rice grain and rice husk are respectively 20.1, 30.2 and 29, 36 in exposed and control site (Fig. 28b). Chowdhury et al. (2020a, b) observed similar findings where translocation % of As in the final ripening phase from root to grain was lower in exposed fields (0.0027%) in comparison to the control fields (0.0215%). This is probably caused by the As stress that infuse lower rate of translocation in the exposed sites

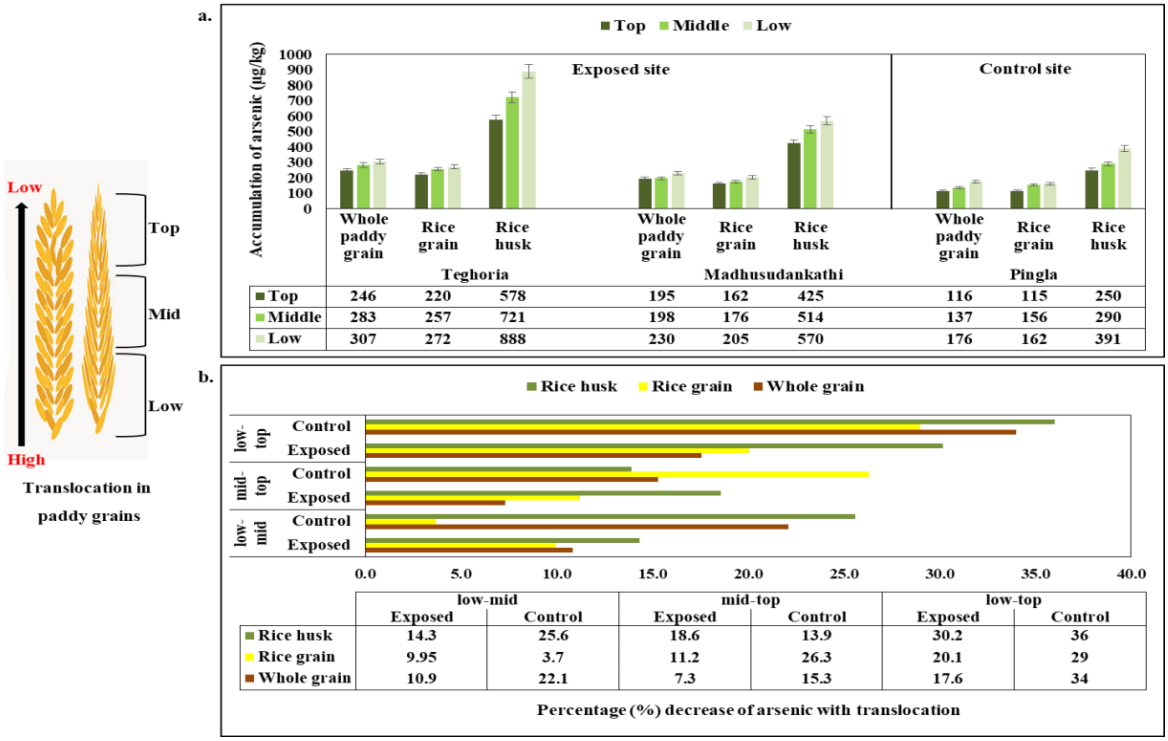


Fig. 28. Translocation of arsenic in different parts of whole paddy grain located in single pedicel (height wise) and percentage (%) decrease of arsenic with translocation

5.3.2. Localization of arsenic in single rice grain

Rice grain As concentration variation is previously discoursed in many articles (Chowdhury et al., 2018a, 2020b) with focus on cultivars and background water and soil concentration. According to the theory of translocation, rice grain accumulates least quantity of As in paddy plant. Concentration of As in the rice system is highest in husk followed by whole grain and grain (Chowdhury et al., 2018b). Apart from estimation of human health risk from As-contaminated rice grain, assessment of As toxicity is required too in livestock as As-contaminated rice husk is a prime part of their regime.

5.3.2.1. Arsenic exposed paddy fields

Cultivars of rice grains (n= 7) were collected from North 24 Parganas district, an endemic site of West Bengal that showed similar pattern of metal accumulation (**Fig. 29a**). In pre-monsoon, Ranjit cultivar grainhad highest accumulation (649 µg/kg) while Masuri showed least (278 µg/kg). Inpost monsoon, Minikit showed highest As accumulation (549 µg/kg) and Maharaj showed lowest accumulation (226 µg/kg) among the studied five cultivars. The average rice As concentration was undoubtedly higher in pre-monsoon than post monsoon (445 µg/kg and 325 µg/kg respectively). However, all the cultivars accumulate

higher As than the maximum tolerable i-As concentration n rice (100µg/kg) (Meharg et al., 2006).

On the other hand, when individual grains are studied, it shows that in most cases, rice grains have higher As content than its husk. **Fig. 29b** demonstrates that in pre-monsoon, rice cultivars Ranjit, Ganga, Sanat Maharaj, Sabsarna and Swarnamasuri show As content in the array of whole paddy grain > rice grain > rice husk. Average dry weight of an individual whole grain, rice grain and rice husk are respectively 0.0223 g, 0.0196 g and 0.0044 g which represents that metal content accumulation is directly proportional to the weight of the subject. Despite the fact cumulative dry weights of a single rice grain and rice husk nearly equals to that of a single whole paddy grain, As content picture is not identical. The difference of As content between a whole paddy grain and a rice grain along with its husk is ranged as (-) 10.9 to (+) 7.8 ng (average: -2.1 ng). The proportion of As content between rice grain and rice husk (range: 0.61-1.90; average: 1.16) seems to be more than that of their As concentration (average: 0.27, range: 0.12-0.45). In post monsoon, about 80 % rice grains followed the above-mentioned order of As content. The fraction of As content between rice grain and rice husk (average: 1.26, range: 0.66-1.75) is observed to be more than that of their As concentration (average: 0.33, range: 0.29-0.44). The range of difference of the As content between a whole paddy grain and a rice grain along with its husk is measured to be (-) 8.3 ng to (-) 0.6 ng (average: -2.4 ng). The pre-monsoonal and post monsoonal average As content of rice grain are 7.8 and 6.54 while that of rice husk are7.9 and 5.66 respectively. This explains the positive effect of rainwater in decreasing As stress in paddy systems (Shrivastava et al., 2017). Monsoonal rice counted less Asthan pre-monsoon cultivation even in As exposed fields. Though, regression analysis reveals that both the As content and concentration between rice grain and husk are well harmonized in two seasons (R^2 values = 0.87 and 0.83; $p < 0.05$). According to Bhattacharya et al. (2010), significant correlation has been noticed too between rice grain and its husk in both Boro and Aman rice (r= 0.959, 0.923). Therefore, As is confined towards the inner side of a grain more and the external husk contains As through adsorption in surface. Yet, Meharg et al. (2008) showed that in white rice, As is distributed all over the grain, the majority of which encompasses the endosperm and it is observed to be selectively confined at the grain surface of brown rice similar to the pericarp and aleurone layer. XRF images of this work evidenced that As is largely located in the surface of the rice grains. Although, high As concentration was present in the aleurone and external parts of the endosperm close to the ovular vascular trace in rice grains (Lombi et al., 2009). The localization study of As also divulges that brown rice holds more As than

white/polished rice which is the metal accretion ability of rice bran. As per the report of Sun et al. (2008), rice bran is reported with 10 to 20 folds more As concentration compared to that in bulk grain. Rahman et al. (2007) disclosed maximum As concentrations in the order of rice hull then bran-polish, brown rice, raw rice and lastly polished rice. Unpolished rice grains have higher As than polished rice. Arsenic concentration was also more in brown rice that contains the external coating of rice than white rice, demonstrated by Yim et al. (2017). The polished rice available in the arcade comprises lower As concentration than the full grain as rice embryo has the maximum As concentration (up to 13000 µg/kg) (Kramar et al., 2017).

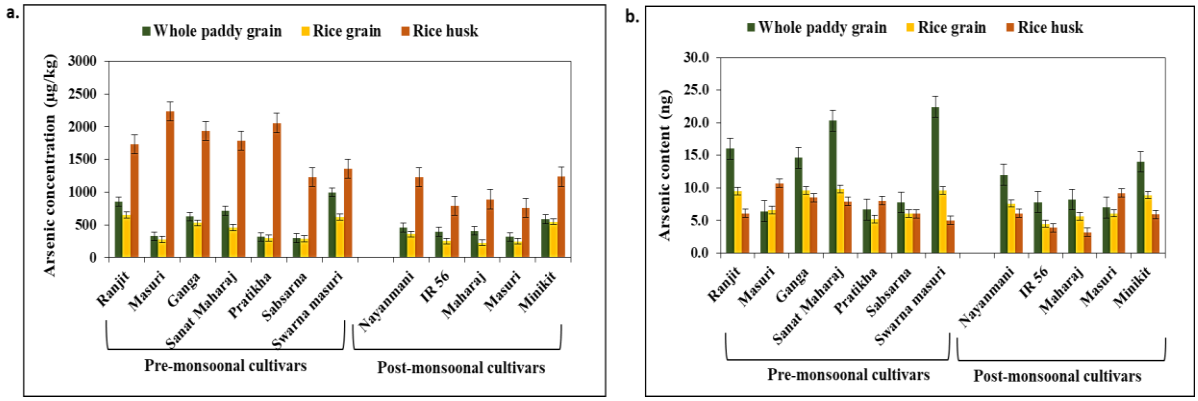


Fig. 29. Variation of arsenic (a) concentration and (b) content in different parts of paddy grain in arsenic exposed fields

5.3.2.2. Arsenic unexposed paddy fields

The same study has been done on As unexposed zone (Pingla) in two different seasonal cultivars. Accumulation of As concentration and content in the rice cultivars have been compared with exposed zone as well as seasonally.

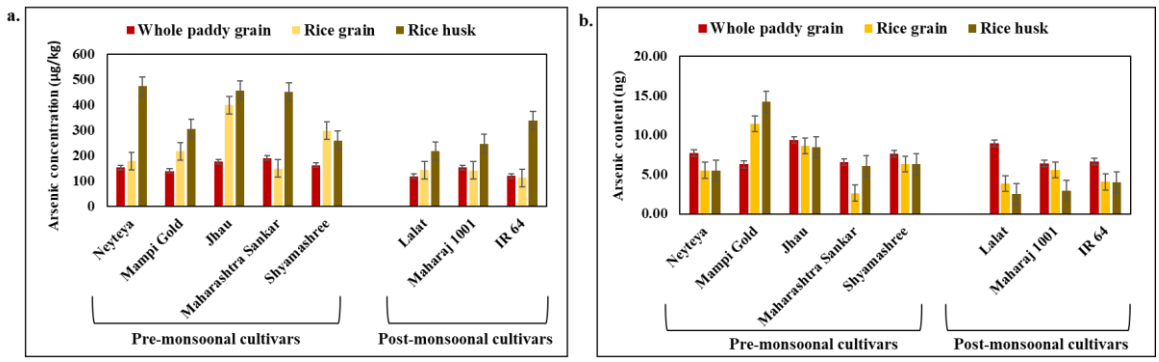


Fig. 30. Variation of arsenic (a) concentration and (b) content in different parts of paddy grain in arsenic unexposed fields

The studied 5 pre-monsoonal rice cultivars and 3 post monsoonal rice cultivars showed substantially lower As concentration than pre-monsoonal accumulation (**Fig. 30**). Highest and lowest As accumulation was observed in Jhau (400 µg/kg) and Maharashtra sankar (149 µg/kg) in pre monsoon whereas the 3 monsoonal cultivars showed similar kind of accumulation; 143, 142 and 112 µg/kg for Lalat, Maharaj 1001 and IR 64 respectively (Fig. 30a). The translocation theory in As concentration was maintained in almost all the cultivars. When single grain As content was studied in rice cultivars, it was seen that in 75 % studied cultivars (three out of three monsoonal cultivars and 3 out of 5 pre-monsoonal), As content was lowest in rice husk followed by rice grain and whole paddy grain. The range of difference of the As content between a whole paddy grain and a rice grain along with its husk is measured to be (-) 19.4 to (+) 2.58 ng (average: - 4.81 ng). The ratio of As content between rice grain and rice husk (average: 1.09, range: 0.43-1.9) is found to be more than that of the ratio of their As concentration (average: 0.62, range: 0.33-1.14). The pre-monsoonal average content of rice grain and rice husk are 6.93 and 8.12 while the post monsoonal average content of rice grain and rice husk are 4.5 and 3.16 respectively.

5.3.2.3. Comparative remarks

From the content analysis, the most important finding is that in the As exposed fields, content of As in a single rice grain is approximately 16.2% less during post monsoonal cultivation than pre monsoon. Likewise, As content in a single rice grain is approximately 35% less during post monsoonal cultivation than pre monsoon in As unexposed or control fields. Cumulatively, it can be concluded that rice grains As content is about 25.6% less in post monsoon compared to grains cultivated in pre monsoon. It is revealed at the same time that in pre monsoon the As accumulation in single rice grain is almost 11.2% less in control area than exposed area, while that is approximately 31.2% less in post monsoon. Monsoonal rice As concentration (average) in exposed fields and control fields are nearly 1.37 and 1.88 times less than pre monsoonal rice grains. Chowdhury et al. (2020b) observed the accumulation of As in rice grain was roughly 3 times lower during monsoonal cultivation than pre-monsoon season in As exposed fields. Pre-monsoonal rice husk Asconcentrations (average) are nearly 1.8 and 1.46 times higher in exposed fields and control fields than post monsoon. Whereas, pre-monsoonal rice husk average As content are about 1.39 and 2.6 times higher in exposed fields and control fields than post monsoon.

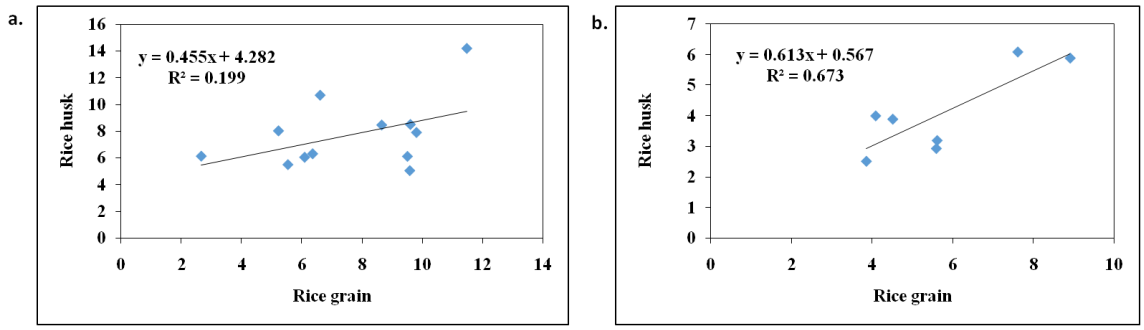


Fig. 31. Regression analysis between rice grain and rice husk arsenic content in (a) pre-monsoon and (b) post monsoon

The association between rice grain and rice husk As content is quite strong during post monsoonal cultivation (R^2= 0.673) (**Fig. 31b**) than pre monsoonal cultivation (R^2= 0.199) (**Fig. 31a**). It is proved that even control sites contributed some amounts of As in monsoonal rice grain in comparison with higher accumulation from exposed sites. It expresses that the monsoonal effect of As reduction in soil paddy system is stronger in unexposed or control sites than the Asexposed fields. It can be explained by the background soil and irrigational water As concentration in the location.

5.3.3. Distribution of arsenic species in rice grain

Arsenic species in rice is mainly in inorganic form accompanied by its methylated species. Dimethyl-arsinic acid (DMA) in a flexible amount and little amount of monomethyl-arsonic acid (MMA) were also present as per Norton et al. (2009) and Williams et al. (2005), (2006). Metabolism of As and the speciation within the paddy plant with special emphasis to rice grain is still booming (Zhao et al., 2009). Report said that the assimilation rate of i-As in paddy plant is much greater than the methylated species (DMA or MMA) (Abedin et al., 2002). As (V) is converted into As (III) in the paddy root system and translocated to paddy grain by xylem through arsenite effluxer. Further reports showed that, arsenite may be depolluted by the development of thiol rich peptides, including phytochelatins (PCs) and glutathione into the vacuoles (Zhao et al., 2009). Methylated species of As have much relaxed invasion than the inorganic species in root system, but they appear in plant systems more adeptly (Raab et al., 2007a). It occurs during translocation because of the less quantity of MMA and DMA along with their lower

impedance ability (Raab et al., 2007b). Rice grain As speciation is dominated by i-As and DMA (Williams et al., 2005). But translocation of DMA was weightier in rice grain than i-As, described by Carey et al., (2010). Inorganic As was principally found in 64 to 81% in the samples of Bangladesh and India, where DMA occurred in very little amount (Williams et al., 2005). A whole plant shows a lower concentration of DMA in rice shoots but surprisingly rice grain shows higher quantity of DMA, reported in Abedin et al. (2002). DMA is more transportable in paddy system than i-As via xylem and phloem tissues (Lombi et al., 2009; Tanaka et al., 2007).

Quantification of As species distribution in raw rice grain and respective rice husk in different farming season and different As exposure zones is given in **Table 25**.

Table 25: Arsenic species distribution in raw rice grain and corresponding rice husk in pre and post monsoon

Cultivation area	Cultivation season	Name of cultivar	Component	Total As (µg/kg)	As III (µg/kg)	As V (µg/kg)	DMA (µg/kg)	MMA (µg/kg)	inorganic As (III + V) %
Exposed	**Pre-monsoon**	Sanat Maharaj	Rice grain	194	100.3059	75.3314	17.8728	-	90.7
			Rice husk	503	41.9319	414.723	46.241	-	90.8
Control		Shayamashree	Rice grain	53.5	29.6915	23.8227	-	-	100
			Rice husk	78.6	9.15969	69.4333	-	-	99.9
Exposed	**Post monsoon**	Ganga	Rice grain	205	68.646	51.5399	85.2569	-	58.6
			Rice husk	922	52.2909	831.726	38.0272	-	95.9
Control		Lalat	Rice grain	36	21.669	14.3322	-	-	100
			Rice husk	113	14.763	98.0802	-	-	99.8

The HPLC chromatograms of standard 25 ppb (µg/L) As mixture solution and rice grains are shown in **Fig. 32**.

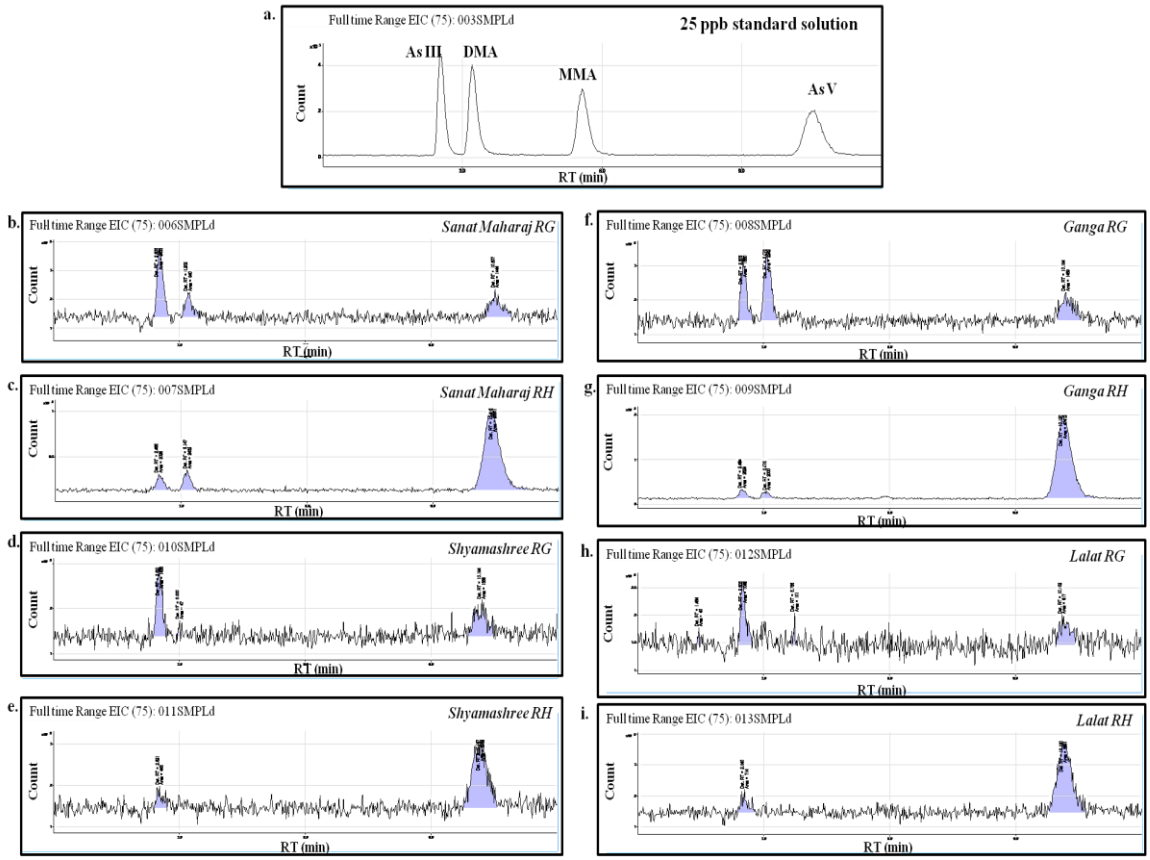


Fig. 32. Chromatograms of arsenic concentrations in rice grain and rice husk of different cultivars: (a) Standard 25 ppb solution, (b) Sanat Maharaj rice grain, (c) Sanat Maharaj rice husk, (d) Shyamashree rice grain, (e) Shyamashree rice husk, (f) Ganga rice grain, (g) Ganga rice husk, (h) Lalat rice grain, (i) Lalat Rice husk

The total Asconcentration in the rice grains cultivated in the Asexposed fields in two different seasons are more or less similar; 194 and 205 µg/kg. But the total As concentration in the rice husk is higher in Ganga cultivar (922 µg/kg) which is cultivated in post monsoon in As exposed fields. This can be explained by the fact that besides water As concentration, As accumulation in husk depends on cultivars and background soil As concentration. In rice grains, the contribution of inorganic arsenic (iAs) from total As is seen to be an average of 87.3 % (range: 58.6 to 100 %) which is in good agreement with the findings of Halder et al. (2013), (2014); Laparra et al. (2005); Meharg et al. (2009); Mondal and Polya (2008) and Signes-Pastor et al. (2008). They showed that the main species of As in rice grain is toxic and carcinogenic inorganic which contributes about 80% of total Ascontent in rice grain. Moreover, the accumulation of i-As from total Asin rice husk in the present study is observed even higher (average: 96.6%; range: 90.8 to 99.9%). In both pre and post monsoon season it has been seen that presence of toxic As-

III is dominant over the presence of less toxic As-V, whereas, the trend is just the opposite in rice husk. Concentration of arsenite (As-III) is higher in rice grain by almost 25.86% (range: 19.76-33.86%) than arsenate (As-V), while in rice husk, presence of As-V is higher than As-III by about 88.8% (range: 84.9-93.7%). I-As concentration is higher in pre monsoonal rice grains by approximately an average of 32 % than the post monsoon season but presence of organic As (DMA) is almost 79% more in post monsoonal rice grains compared to pre-monsoonal rice. This corroborates with the main findings of Chowdhury et al. (2020b) that rice grains cultivated during post monsoon are comparatively safer for consumption than the pre-monsoonal rice. Similarly, presence of i-As is higher in the rice grains cultivated in As exposed areas than the unexposed or control fields by approximately 69.7%. Presence of MMA is found in none of the rice grain and husk. It is evident that i-As mainly dominates grain As concentration with presence of less amount of DMA (Halder et al., 2014). Although, i-As concentration in rice husk is drastically opposite; nearly 61.6% more in post monsoonthan the pre monsoon season but presence of organic arsenic (DMA) is almost 17.8% less in post monsoonal rice husk compared to pre-monsoonal. Arsenic content in single rice grain is also 25% decreased in monsoonal cultivation than the pre-monsoonal cultivation. This is well described in **Fig. 33**.

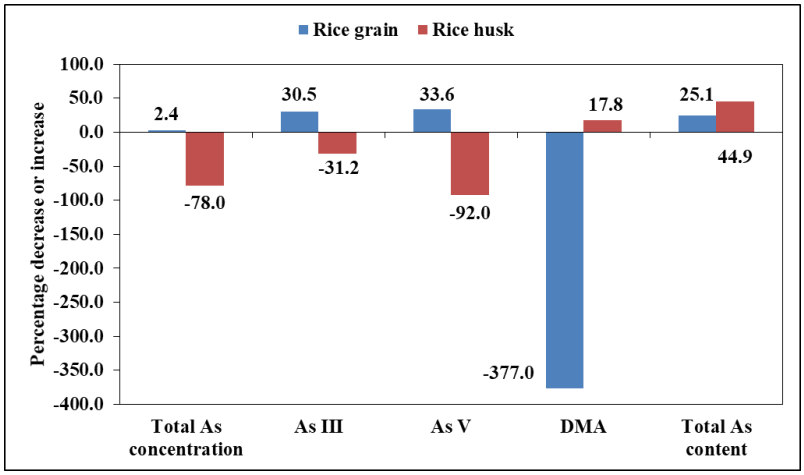


Fig. 33. Variation of arsenic species concentration in rice grain and rice husk

5.3.4. Interpretation of flow of arsenic during cooked rice production

Toxicity of As is enlarged by consuming contaminated cooked rice. The movement of As quotient between As-contaminated rice and cooking water is complex. Assimilation of As in cooked rice is dependent on various factors like rice variety, cultivar, As concentration in raw rice and cooking water, cooking practice, cooking utensils etc. (Halder et al., 2012,

2014; Rahman et al., 2011; Sengupta et al., 2006). The existence of micronutrients or other minerals may also impact cooked rice As concentration or As species dispersal, showed in Batista et al. (2011), Chowdhury et al. (2020a), Mwale et al. (2018). When rice is prepared with low (<3 µg/L) to moderately As contaminated water (36 to 58 µg/L), As discharges from cooked rice to water but when prepared with higher than 80 µg/L As concentrated water, As is added in cooked rice from contaminated water (**Fig. 34a**) (Chowdhury et al., 2020a). This study stated that sustaining an optimum As concentration in cooking water is necessary constantly to outline the movement of As between rice and water. But, no fixed law was followed in movement of As for different varieties of sunned and parboiled rice grain. Furthermore, some rice cultivar like Nayanmoni, Saatswarna and Minikit disclosed upper threshold water As accumulation value during cooking (Chowdhury et al., 2020a).

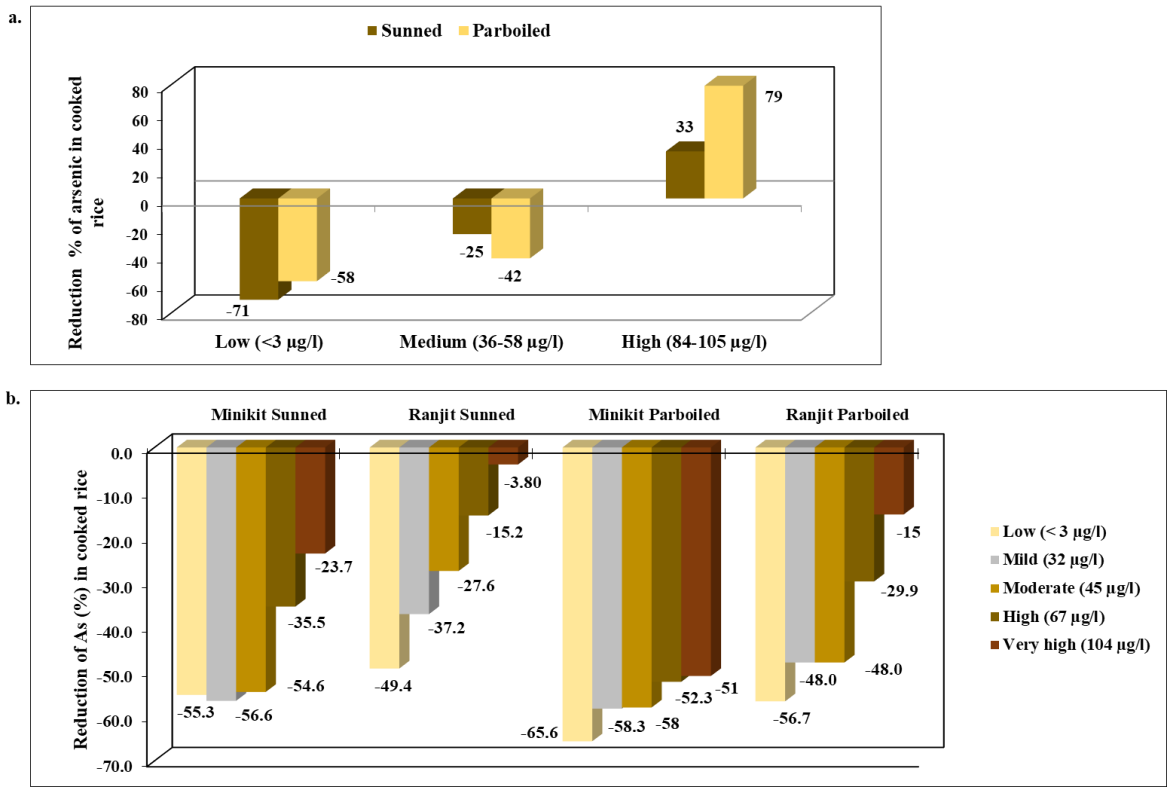


Fig. 34. Flow of arsenic between rice grain and water: a) Threshold value of water arsenic accumulation during cooking according to Chowdhury et al., 2020a, b) Release of arsenic from cooked rice (with respect to cultivar and variety) during cooking with five different As-concentrated water.

The existing study therefore explored the movement of As while cooking considering definite rice variety and cultivar. Both the sunned and parboiled varieties of rice in Minikit

and Ranjit cultivars were prepared with 5 different As concentrations of water that are <3, 32, 45, 67 & 104 µg/L. It revealed that at the time of cooking by the differently As concentrated waters, As discharged from cooked rice to water; however the degree of discharge got lessened with increasing water As concentration (**Fig. 34b**). It was found that highest As (%) was discharged from raw grain to cooked rice (55.3, 49.4, 65.6 and 56.7) when cooked with As= <3 µg/L and lowest As (%) (23.7, 3.80, 51 and 15) was discharged with As= 104 µg/L water As concentration in Minikit sunned, Ranjit sunned, Minikit parboiled and Ranjit parboiled correspondingly. Hence, the cooking water As concentration and As decrease % in cooked rice appears to be in reverse. The average As release aptitudes of Minikit cultivar were 60.4, 57.5, 56.3, 43.9 and 37.4% during cooking, which were relatively greater than the Ranjit cultivar (53, 42.6, 37.8, 22.6 and 9.4%). Likewise, release of As was more in the parboiled varieties of cooked rice (61.2, 53.2, 53.1, 41.1 and 33%) than the cooked sunned variety (52. 3, 46.9, 41.1, 25.4 and 13.7%). It can be elucidated through the detail that the raw rice grain accrues As during a the boiling process of parboiling (Chowdhury et al., 2018b) which may be inaccurately bound over the external part of rice and effortlessly discharged during cooking. Summative, Minikit parboiled rice grains had highest potential of As discharge during cooking with As concentration <3-104 µg/L. This study also foundgreaterAs reduction % with higher As concentration in raw rice grains. In Minikit sunned, Minikit parboiled, Ranjit sunned and Ranjit parboiled rice As discharge % from corresponding cooked rice was 45, 57, 26.6 and 39.5 whereas the initial As concentration in these raw rice grains were respectively 152, 384, 79 and 127 µg/kg (**Fig. 35a**). However, Chowdhury et al. (2020a) observed that with greater initial As concentration in raw rice, the increase % of As in cooked rice was less even if the rice is made with As-contaminated water. The current study also noticed that the As concentration in the gruel (discarded water) amplified with increasing cooking water As concentration (**Fig. 35b**). The maximum As concentration was observed in each gruel sample (95, 182, 68.6 and 96 µg/kg) while cooked with As = 104 µg/L. It bared that water As concentration holds a substantial influence on As concentration in cooked rice and gruel.

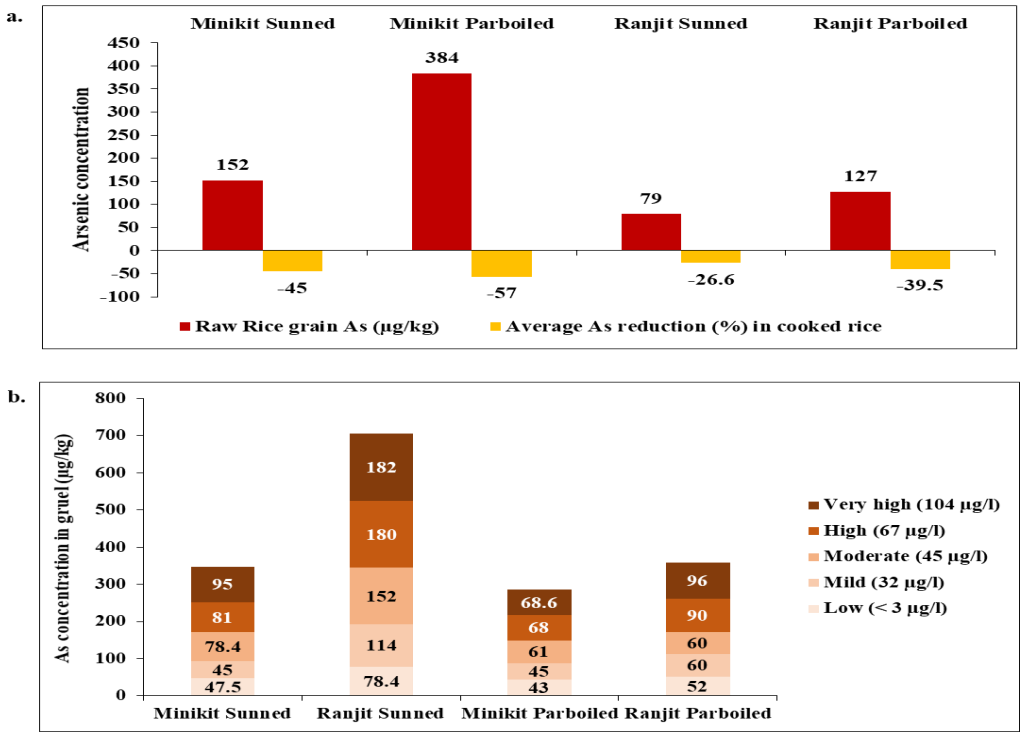
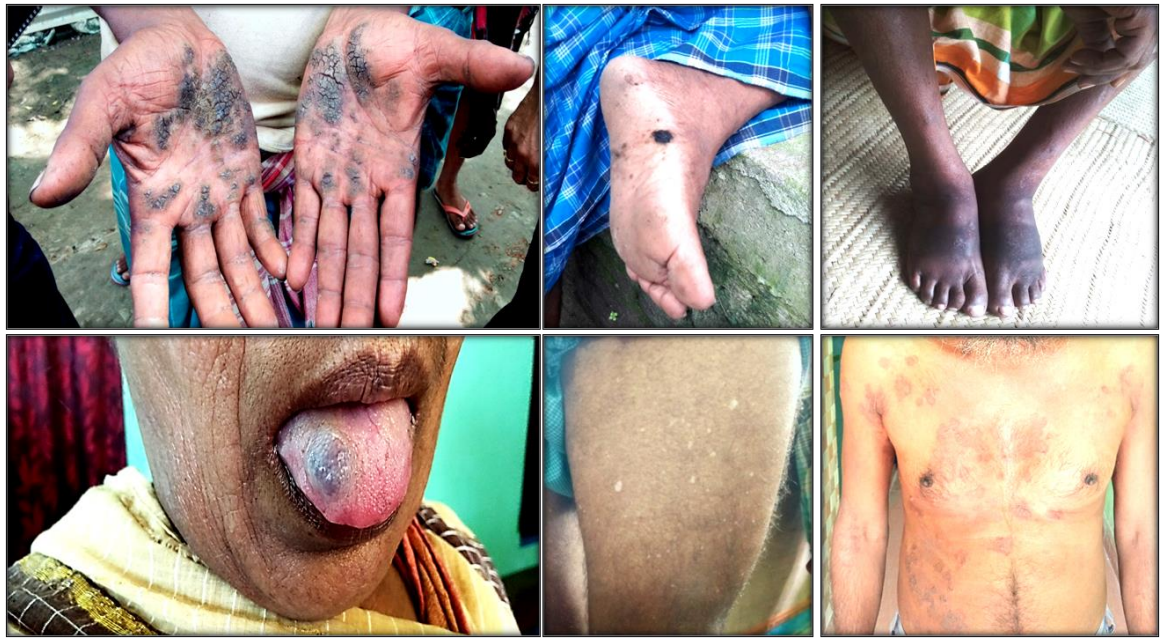


Fig. 35. Flow of arsenic during cooking: a) with respect to raw rice grain arsenic concentration, b) gruel arsenic concentration

Arsenic contamination in rice grain is one of the most disturbing topics recently. It is required to remove As from the origin to reduce As concentration in rice, which is quite challenging. Therefore, the rural As exposed people are advised to cook rice with As safe or As free water.

5.4. Effect of arsenic toxicity on humans



Extent of As exposure and its associated future health risk to the population of different areas in West Bengal has been assessed under the present research study based on several hypotheses. The hypotheses have been tested on some proposed explanation for few observed phenomena and performed on quantitative research data.

5.4.1. Assessment of the quality of health in studied school children from arsenic exposed areas of 'Nadia'

Two different age groups of school children (9-10 years and 14-15 years) studying in Laupala Kalpataru Primary School, Jaguli Junor Basic School and Laupala Kalpataru High School from Chakdah and Haringhata Municipality area under the jurisdiction of Nadia district have been selected as the subject of the study. The history of As exposure in the studied area has been explained earlier.

5.4.1.1. Drinking water arsenic contamination status

The study initiated when drinking water samples of the 15 schools of Haringhata town were analyzed and showed As concentration in a range between 60.2 and 366 µg/L with a mean value of (136 ± 79) µg/L. Also, 22 schools were observed to contain tube well water with As concentration > 10 µg/L in Chakdah (average: 60 µg/L; range: 11.4 to 219 µg/L). Previous research by Rahman et al. (2014) in Tehatta II block of Nadia district showed that 87.5% of drinking water samples of school premises (total sample n= 96) were beyond recommendation with a maximum of 254 µg/L. In the present study, three school tube-wells that have highest As concentration among the studied samples were selected. The drinking water source of Laupala primary school showed As concentration of 216 µg/L which is being used by approximately 280 persons everyday whereas that from the Jaguli Junior Basic school and Laupala high school was 219 and 366 µg/L and being used by around 700 and 850 persons daily. This observation pushed us to undertake the present study i.e. evaluation of health exposure in selected children of the three selected schools distinguished by two different age groups (9-10 years and 14-15 years).

Besides drinking As contaminated water during school time, analysis of domestic drinking water samples of the two groups of children showed that they have been still consuming As contaminated water knowingly or unknowingly in their houses. As earlier described, Nadia being an As affected district, several mitigation strategies have been undertaken till date. Still, the ill effect of Ascontaminated drinking water and the sufferings of poor affected people is unbearable in Nadia (Chakraborti et al., 2013, Rahman et al., 2014). The

mean As concentrations in the domestic drinking water of the two groups of children are observed to be 44.2 ± 47.2 and 45.5 ± 53.6 µg/L (range: <3-248 µg/L; n= 93 and 3-210 µg/L; n= 50) respectively (**Table 26**).

Table 26: Drinking water and rice grain arsenic concentration status from the children's houses

Age group: 9-10 years						
Parameters		**No. of samples (n)**	**Mean**	**Median**	**SD**	**Range**
Domestic drinking water (µg/L)	**Chakdah (Laupala Kalpataru Primary school)**	45	25.4	14.9	26	3-101
	Haringhata (Jaguly Junior Basic School)	48	65	56.9	56.2	<3 - 248
Total		93	44.2	28	47.2	<3-248
Rice grain (µg/kg)	**Chakdah (Laupala Kalpataru Primary school)**	45	186	168	78	89-354
	Haringhata (Jaguly Junior Basic School)	42	288	270	140	111 - 900
Total		97	202	217	117	89 - 900
Age group: 14-15 years						
Chakdah (Laupala Kalpataru High school)	**Domestic drinking water (µg/L)**	50	45.5	18.5	53.6	3-210
	Rice grain (µg/kg)	45	180	160	122	39-650

5.4.1.2. Rice grain As contamination status

Rice, being a staple food in West Bengal, 73% of total calorie intake in an individual is contributed by it (Ninno and Dorosh, 2001). But, with time it has been systematically proved that rice is a major route of As exposure to the mankind. Paddy plant is a huge accumulator of As and consequently rice grain is a serious health risk factor to be taken care of (Chowdhury et al., 2018a, Chowdhury et al., 2020a, 2020b). Paddy cultivated in the fields of Nadia district is sufficiently As contaminated and much efficient to cause health risk in future (Mondal and Polya, 2008, Roychowdhury, 2008). Rice grain cultivated in Chakdaha block, Nadia showed As accumulation of 950, 790, 600, 470 and

290 µg/kg in Gosai, Satabdi, Banskathi, Kunti and Ranjit variety respectively (Upadhyay et al., 2019). The present study analyzed As concentration in the rice grain samples consumed by the children in a daily basis. It showed that the range of As concentration in the lower aged group of children was 89 – 900 µg/kg (n= 97) while that in the higher aged group was 39-650 µg/kg (n= 45), though the mean As concentration was higher in the former one (232 ± 117 and 180 ± 122 µg/kg). In both the cases, the rice grain As concentration is higher than the allowable As concentration in rice i.e. 100 µg/kg of As endemic sites (Meharg et al., 2006).

5.4.1.3. Daily dietary intake of As

Daily dietary intake of As is calculated on the basis of the daily consumption of rice and water for the studied groups of children as in the Bengal delta, people mostly rely on rice and vegetables. Raw rice, parboiled rice and different types of rice by products are enough As contaminated to cause health risk for the population residing in the As exposed area (Chowdhury et al., 2018b, Islam et al., 2017). Groundwater used for drinking and rice grain both contain ample amount of inorganic As and more importantly, above 80% of the total As content of food is reflected from inorganic As (Huq and Naidu, 2003, Roychowdhury et al., 2010, Roychowdhury, 2008; Signes-Pastor et al., 2008). Presently, the provisional tolerable daily intake (PTDI) value of inorganic As is considered as 3.0 µg/kg bw/day, based on the range of 2–7 µg/ kg bw/day (WHO, 2011). Our study reveals that in the present study area, the daily dietary intake of both the studied age group of children is way much higher than the recommended one **(Table 27)**. Dietary intake of As for the elder group of children (14-15 years) is 7.82 µg/kg bw/day (range: 5.10 – 13.69 µg/kg bw/day) which is comparatively similar to the younger group of children (9-10 years), 7.99 µg/kg bw/day (range: 4.08 – 13.32 µg/kg bw/day). It is to notice that As concentration of drinking water from school tube-well contributes maximum towards the daily As intake among the three sources (3.63 and 4.57 µg/kg bw/day respectively). Surprisingly, rice grain As concentration is a potent factor towards the daily body burden of As. Rice grain As contributes 2.93 and 1.71 µg/kg bw/day respectively in the younger and elder groups of children which signifies that body weight is a less significant factor than the As concentration.

Table 27: Dietary intake of Arsenic per day in the two studied groups of children

Source	Consumption rate per day*		As concentration (ppb)		Daily dietary intake of As per day (µg/kg bw/day)**	
	9-10 years	14-15 years	9-10 years	14-15 years	9-10 years	14-15 years
School tube-well drinking water	0.5 l	0.5 l	217.5 (average of 216 and 219)	366	3.63	4.57
Domestic tube-well drinking water	1.5 l	2 l	44.2	45.5	2.20 (range: 0.04 – 12.3)	1.70 (range: 0.11 – 7.9)
Rice grain consumed in home	0.38 kg	0.38 kg	202	180	2.93 (range: 0.93 – 11.4)	1.71 (range: 0.37 – 6.17)
Total	-	-	-	-	7.99 (range: 4.08 – 13.3)	7.82 (range: 5.10 – 13.7)

*Joardar et al., (2021b);**$BW_{(9-10)}$ = 30 kg, $BW_{(14-15)}$= 40 kg

5.4.1.4. Biomarkers: Health Exposure

The health exposure study of the children is studied through analysis of their biological samples i.e. urine, hair and nail and elaborated in **Table 28**. Urine is the measurement of acute exposure as As is primarily metabolized in liver and most of its species are excreted through this pathway (Buchet et al., 1981, NRC, 1999, Orloff et al., 2009). The average urine As concentration in the present studied children (9-10 years: 4.91 µg/L, 14-15 years: 5.49 µg/L) is found to be strictly within normal range i.e. 3-26 µg/L (Joardar et al., 2021b). The urine As concentration range in the two groups are (<3 – 15.4) and (<3 – 18.5) µg/L respectively; (n= 89 and 40). Moreover, reports say that individuals not exposed to As may have urine As below 100 µg/L (ATSDR, 2007, Chakraborty et al., 2016). After urine As excretion in the first one or two days of direct exposure, As gets slowly accumulated in hair. Therefore, hair As can speak about long time or past exposure as it reflects accumulation which growing with age (Orloff et al., 2009). The Normal range of hair As concentration is 80–250 µg/kg (Arnold et al., 1990) while 20- 200 µg/kg is stated as usual As concentration range in hair of the people with no As exposure (NRC, 1999). Hair As is mostly arsenite [As (III)], though a few researchers mentioned the existence of DMA in hair (Lin et al., 1988). The present studied population has seemingly higher As concentration in hair which shows that they are exposed to As for a stretched period of time (Table 28). The scalp hair As concentration range in the elder children is 400-13200

µg/kg (n= 45) while that in the lower aged children is 100 - 6400 µg/kg (n= 90). Finger and toenails are intermittently used as biomarker for As exposure study as it grows at a slower rate than hair (Orloff et al., 2009). According to National Research Commission, normal range of As in nails of people having no As exposure is 20-500 µg/kg (NRC 1999, 2001) while its referred range is stated as 430–1080 µg/kg by Ioanid et al. (1961). Nail As concentration range in the present studied population has also crossed the referred limits; 120-9300 µg/kg in the elder group of children and 410-8230 µg/kg in the younger one.

Table 28: Status of the biological samples in children

Parameters	Biomarkers					
	Urine As (µg/L)		Scalp Hair As (µg/kg)		Nail As (µg/kg)	
	9-10 years	14-15 years	9-10 years	14-15 years	9-10 years	14-15 years
No. of samples (n)	89	40	90	43	93	44
Mean	4.91	5.49	1607	1240	2174	2400
SD	2.70	3.26	1143	2550	1516	2140
Range	<3 – 15.4	<3-18.5	100-6400	400-13200	410-8230	120-9300

5.4.1.5. Relation between daily dietary intake of As and excreted As through biomarkers

A statistical performance named 'two-tailed paired t test' has been done at 95% confidence level with hypothesized mean difference zero to identify the relationship between daily dietary As intake and the As excreted through the biomarkers. Null hypothesis (Ho) is considered as 'no significant difference exists between DI and As concentration in biomarkers' and alternate hypothesis (H_1) is considered as 'significant interdependence between DI and As concentration in biomarkers'. The t test values in each relation with respective degrees of freedom (df) are given in **Table 29**. It is statistically proved that there is a significant association of daily As consumed through diet (drinking water and rice grain) with As accumulation in the body markers. The association among Asconcentration in drinking water and As concentration in biomarkers had been also established by Joardar et al. (2021a) by single factor ANOVA.

Table 29: 'Two-tailed paired t test' analysis for estimation of the dependence between DI (dietary intake) of arsenic and arsenic concentration in biomarkers

Age group		DI – Urine As	DI – Hair As	DI – Nail As
9 – 10 years	df	82	83	87
	t test	$t_{stat} > t_{critical\ (two\ tailed)}$; (9.16 > 1.99)	$t_{stat} > t_{critical\ (two\ tailed)}$; (22.3 > 1.99)	$t_{stat} > t_{critical\ (two\ tailed)}$; (15.5 > 1.99)
	Remarks	H_o rejected	H_o rejected	H_o rejected
14 – 15 years	df	41	42	43
	t test	$t_{stat} > t_{critical\ (two\ tailed)}$;(4.41 > 2.02)	$t_{stat} > t_{critical\ (two\ tailed)}$;(13.2 > 2.02)	$t_{stat} > t_{critical\ (two\ tailed)}$; (11.9 > 2.02)
	Remarks	H_o rejected	H_o rejected	H_o rejected

5.4.1.6. Future health risk in the studied children

Health risk assessment is a very important and necessary study towards the estimation of extent of exposure and future health hazard. Risk of As induced diseases increases with increasing toxicity in body system, that too with increasing As intake. In an As exposed and endemic area like Haringhata, it is mandatory to evaluate the health risk of the children who are apparently consuming As a daily basis. Health risk is calculated in accordance of the lifetime cancer and non cancer risk model set by USEPA (1996, 2001). At first, average daily dose (ADD) of As due to consumption of As contaminated drinking water and rice grain is calculated (eq. 9 -11). Where, C = As concentration in respective item, IR = ingestion rate (L/day for drinking water, g/day for rice grain), ED = Exposure duration (9.5 yrs and 14.5 yrs), EF = Exposure frequency (365 days), BW = Body weight (30 and 40 kg), AT= Average life time (365×65 days). In the present studied population, it is seen that the both group of children face substantial amount of health risk for future, although it is prominent than the elder children have more of it than the younger one, probably because of longer duration of As exposure in their lifetime. Drinking water contributes significant amount of cancer and non-cancer risk compared to rice grain. The health risk of both children group is figuratively explained through **Fig. 36**.

Estimated average cancerous risk for the younger and elder group of children are 3.82×10^{-3} (range: 3.2×10^{-3} to 6.8×10^{-3}) and 8.6×10^{-3} (range: 7.71×10^{-3} to 1.2×10^{-2}) through intake of drinking water As. Non-cancerous risk (HQ) values for both the groups are estimated as 8.49 and 19.12 (range: 7.09 – 15 and 17.15 – 26.8) through drinking water. The $HQ_{drinking\ water}$ value is observed to be about 2.26 times higher for younger aged children

compared to the elder group of children. Through the intake of rice grain As, the estimated average cancerous risk for the younger and elder group of children are 6.43×10^{-4} (range: 2×10^{-4} to 2.49×10^{-3}) and 5.7×10^{-4} (range: 1.24×10^{-4} to 2.06×10^{-3}), whereas, the $HQ_{rice\ grain}$ values are 1.43 (range: 0.46 – 5.55) and 1.27 (range: 0.27-4.59) respectively. The average cancerous and non-cancerous values are clearly more than the threshold value of As induced cancer risk and non-cancer risk, 10^{-6} and 1 respectively (USEPA, 2005). The average values of cumulative cancer risk (summation of drinking water and rice grain) is 4.5×10^{-3} (range: 3.4×10^{-3} – 9.3×10^{-3}) for younger group of children and 9×10^{-3} (range: 7.84×10^{-3} – 1.41×10^{-2}) for older group of children respectively. Estimated cumulative hazard quotient values are very high for the two respective groups; 9.93 (7.55 – 20.6) and 20.4 (17.4 – 31.4). It can be concluded that the influence of rice grain As is apparently low compared to drinking water in estimated health risk. Apparently, the elderly children are having higher health risk than the younger group which is mostly caused by longer exposure duration, higher concentration in food and intake rate. Although no skin manifestations are observed in these children, they are clearly sub-clinically affected.

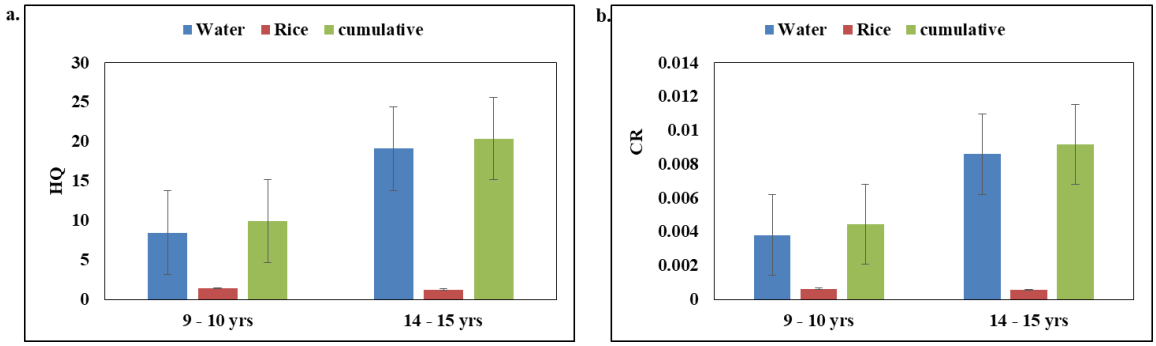


Fig. 36. Cancerous and non-cancerous risk for the studied children

5.4.2. Evaluation of health quality of selected population from Raninagar II, Murshidabad with special reference to age, sex and daily arsenic intake

For reduction of groundwater As contamination in Murshidabad, the government has taken several mitigation measures including installingfresh hand pumps, large diameter deeper tube-wells, setting up As removal plants and supply of pipeline water based on treated surface water etc. Till the year 2007, 80 and 34 dug wells are installed in the affected areas respectively under the 2nd action plan and State sponsored action plan (Halder, 2019). Still, wefound the ineptitude of the governmental mitigation policies in Murshidabad as the pipeline water and As removal plants'treated water are found to be As-contaminated.

Thus, inspection of the situation of the villagers after the administrative involvementstaken in the past years is immediately needed.

The current research targets to measure the level of As exposure to the affected population selected from 60 families comprisingchildren, teenagers and adults, three different age groups residing in Raninagar II block. The work will be done based on three major hypotheses associated toAs intake through drinking water and rice grain, age and gender. The distribution shapeof As in drinking water, urine, hair and nail has been monitored byseparate probability distribution plots or Q-Q plots. Accretion of As in the biological matrix has been estimated throughsex and age variances and the correlationamid As intakes and excretes. Additionally, it is anexclusive statistical effort on finding the most effectual biomarker for As toxicity. Variation in all types of existing drinking water sources have been assembled to outline the most innocuous source of drinking water. Cancerous and non-cancerous risk assessment has been performed extensively on the targeted population via consumption of As-contaminated water and rice. Lastly, another major aim of this study is to inspect the comparative importance of the fundamental factors in calculation of health risk of the three age groups. This has been assessed by Monte Carlo (MC) simulation and a hierarchical cluster analysis (HCA). Drawing the attention of the policy-makers with underlining the present situation after numerous mitigation stratagems taken by the system, is novelty of thepresent work.

5.4.2.1. Arsenic concentration scenario in available drinking water sources and rice grain

Almost 70.6, 60 and 71.4% of drinking water samples were analyzed with Asconcentration more than the approved limit in drinking water in the three affected gram panchayats namely Malibari- I, Katlamari- I and Raninagar- II (**Fig. 37a**). The mean and range of As concentrations in drinking water were observed as 115, 178 and 294 µg/L, respectively and <3-598 µg/L, <3-754 µg/L, <3-847 µg/L, respectively. As a whole, the mean As concentration in drinking water samples of the three surveyedarea is nearly 17.6 folds higher than the WHO (2011a) provided suggested level of As. The population mostly depends on groundwater for drinking; whereas some optional water sources are available too such as treated water from ARPs or pipeline supplied water. The respective mean As concentrations were observed as 208 µg/L (range: <3-847 µg/L, n=35) in the regular tube-

well samples, 54 µg/L (range: 4.1-250 µg/L, n=12) in treated water of Sajaldhara plant and 27 µg/L (range: 3-68 µg/L, n=5) in pipeline water samples (**Fig. 37b**).

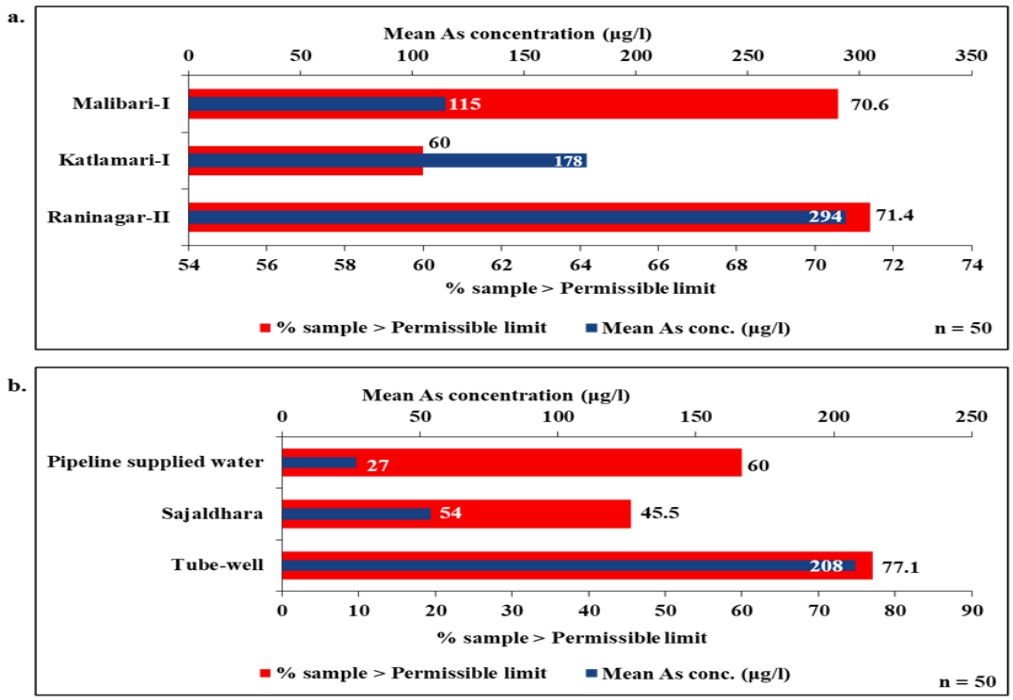


Fig. 37. Drinking water arsenic contamination distribution: (a) in the study area (b)in three different water sources

It was also observed that around 77.1% tube-well drinking water samples, 60%pipeline and 45.5% Sajaldhara plant treated water are As-contaminatedmore than the allowance (**Fig. 37b**). Tapping of pipelines unlawfully for irrigation or any internal purpose is common in thesearea, which eventuallycreateimpairment of the pipes during water supply. It is seen that the basis of accessible pipeline water in few places is only withdrawal of groundwater instead of surface treated water and directly supplied after storing in tanks over-night. Apart from it, the meagreefficacyof As-Fe removal plants in our state compels the inhabitants to have As-contaminated water naively. This clarifies the negligenceof the water treatment plants along with no consistent monitoring of water quality aforeto supplyit to the dwellersresulting into their badhealth. Arsenic mediated health effects are observed pre-clinically and post-clinically. Mandal et al. (2022) and Mukhopadhyay et al. (2020) showed the frequently observed symptoms like diffuse or nodular keratosis, spotted and diffuse melanosis as well as oedema of the feet, enlargement of liver and spleen etc. The distress and misery observed in the community responses are indescribable. The rice grain samples collected from the families showed Asconcentration between 52 to 668

µg/kg (mean: 211 ±167µg/kg). It is significant to discourse that the acceptableAs concentration in rice grain is 200 µg/kg by European Commission (2015). 200 µg/kg has also been advised as the highest i-As concentration in white polished rice which covers 79% of the international market (Codex Alimentarius Commission, 2014). Researchers has thoroughly publicized the effect of As-contaminated irrigational water on rice in Murshidabad. Near about, 0.18% As is accumulated in rice grain each year from deposition in soil that explains the brutality of As exposure in agronomic lands.

5.4.2.2. Daily dietary intake of arsenic in the population

Diverse contributions of dissimilar As-contaminated sources in the daily diets of the different aged populaceswas observed in the present study (**Fig. 38**).

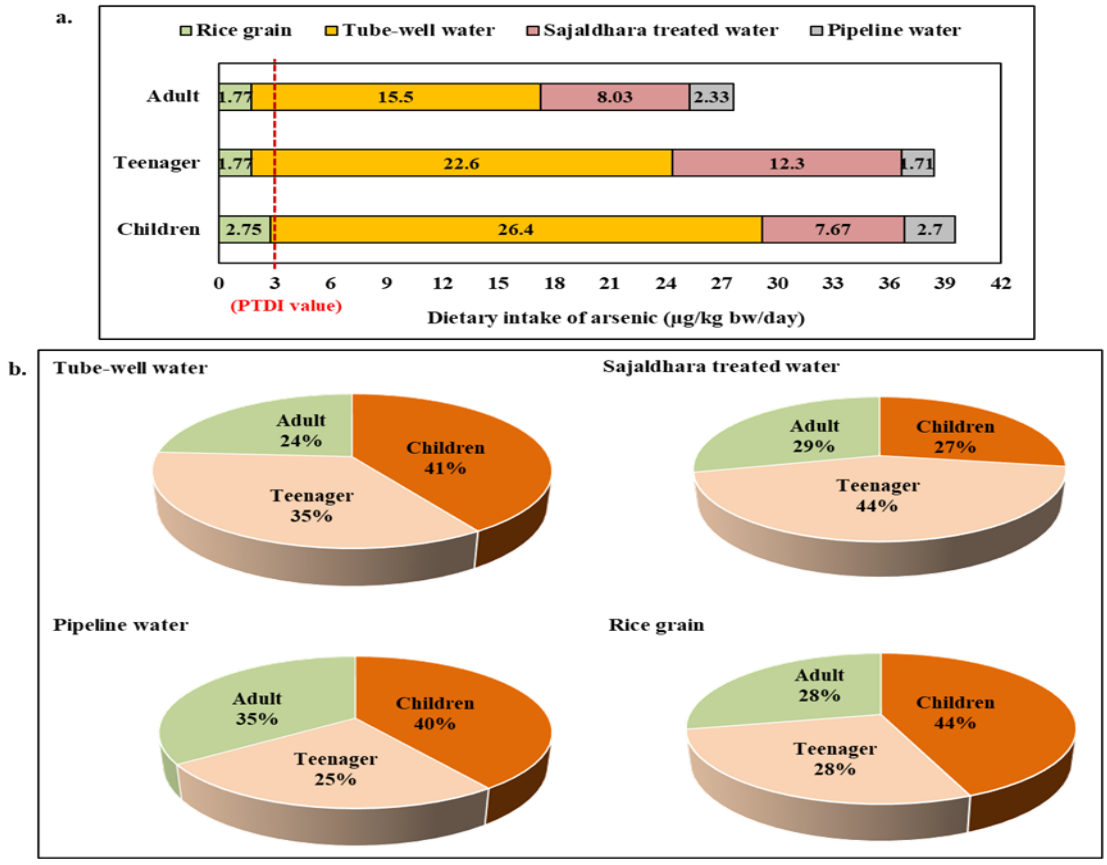


Fig. 38. Daily dietary intake of arsenic: (a) comparison among the three differently aged populations with provisional tolerable daily intake value (b) contribution % of arsenic from different sources

Itfound that bulk of the people rely on water withdrawn through the tube-wells compared to thesubstitute water sources. The results showed that the intake rate of As per day is

highest from tube-well water. The average values are 26.4 (range: 0.31 to 89.2) µg/kg bw/day for children, 22.6 (range: 0.47 to 56.5) µg/kg bw/day for teenagers and 15.5 (range: 0.18-67.8) µg/kg bw/day for adults, respectively. Ingestion rate of As rates in three age groups from the treated water of Sajaldhara plants was 7.67 (range: 0.43 to 43.6), 12.3 (range: 0.48 to 27.6), and 8.03 (range: 0.23 to 33.1) µg/kg bw/day, respectively (Fig. 38a). Average As intake rate per day from pipeline water was found lesser than the PTDI value; 2.7 µg/kg bw/day (range: 0.99 to 4.41), 1.71 µg/kg bw/day (range: 0.63 to 2.79) and 2.33 µg/kg bw/day (range: 0.58 to 3.35) in children, teenagers and adults, respectively. More to it, As-contaminated rice grains also enforced a substantialencumbrance on the daily diets of the community. Ingestion rate of As through rice grains were observed to bemore in children (average: 2.75, range: 1.10 to 5.48 µg/kg bw/day) than teenagers (average: 1.77, range: 1.11 to 2.04 µg/kg bw/day) and adults (average: 1.77, range: 0.32 to 5.34 µg/kg bw/day). The percentage contribution of As in daily foodseems to be maximum in the children group from all the consumption pathways. The dietary As imposed 41, 27, and 40%in children;35, 44 and 25% in teenagers; and 24, 29 and 35%in adults respectivelyfrom tube-well water, Sajaldhara water treatment plant water and pipeline supplied water; given in Fig. 39b. As-contaminated rice grain contributed 44, 28 and 28% in daily dietary intake for the three consecutive groups (Fig. 38b). The variation in data helps to apprehend that the intake rate rests on both‘As concentration in the intake source’ and ‘body weight of the participants’. It also exhibits that drinking water and rice grain subsidize As toxicity of the inhabitants simultaneously.

5.4.2.3. Distribution of arsenic in drinking water and biological indices

Distribution of As in drinking water and the biological indices are extremely skewed, established through corresponding probability distribution diagrams (**Fig. 39**). The Quantile-Quantile or Q-Q plot is a statistical method where the datasets are tested whether they are distributed normally compared to a theoretic normal distribution. Concentration of Asin urine (n=118), hair (n=118) and nail (n=118) samples haveskewness values of 1.66, 1.91 and 1.29 which is positively skewed (Fig. 39b, 39c, 39d).

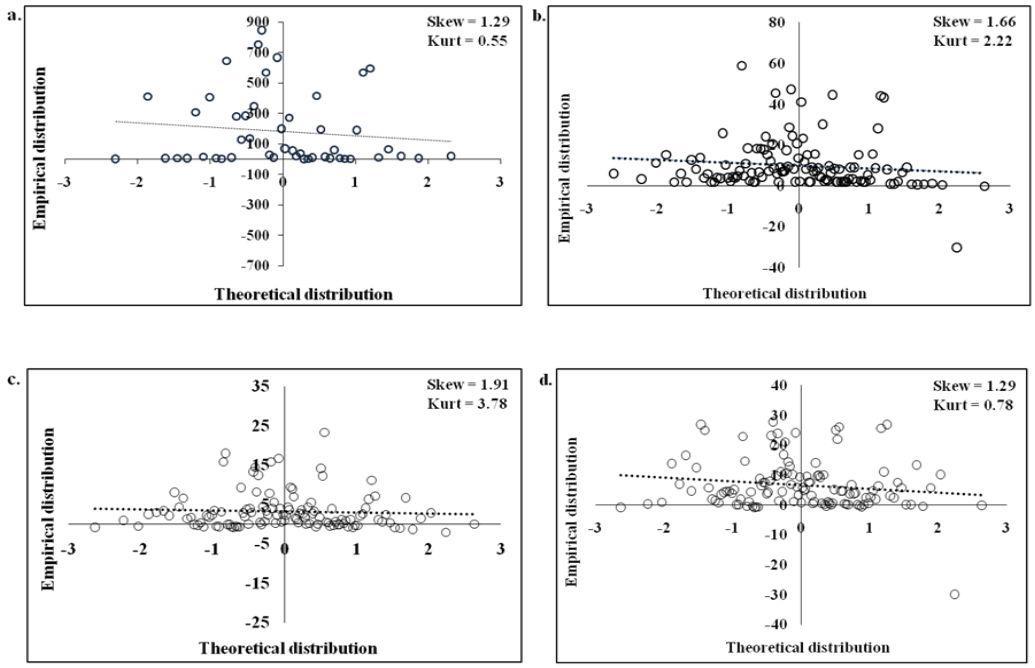


Fig. 39. Probability distribution of arsenic through Q-Q plot: (a) Drinking water (b) Urine (c) Hair (d) Nail

Biological As concentrations were all skewed to the right too in Hinwood et al. (2003). Kurtosis value in a typical normal distribution is 3 and namedas 'mesokurtic'. Distribution of hair As concentration is leptokurtic with a kurtosis value of 3.78 thatmeansthe distribution is heavily tailed with a profusion of outliers. Distribution of urine and nail As concentration are less peaked (platyurtic) with correspondingkurtosis values of 2.22, and 0.78. Concentration of As in drinking water (n=50) is rightly skewed (1.29) and platykurtic with a small kurtosis value (0.55) (Fig. 39a), signifying the distribution is lightly tailed with a deficiency of outliers and comparatively flat. It can therefore be specified that distribution of hair As is least symmetric and extremely peaked.

5.4.2.4. Arsenic concentration in biological samples

The magnitude of As toxicity is usuallyevaluatedbyanalyzing As concentration in human biological samples (urine, hair and nail). Approximately 70% of i-Asgets expelledout from the body system over urine with a half-life of about 4 days (Hughes, 2006). Therefore, urinary As is used as primary body marker for estimation of recent or acute exposure (Buchet et al., 1981; NRC 1999; Vahter et al., 1994). Mean As concentration in urine in children, teenagers, and adults were (11.9 ± 11.3µg/L), (11.1± 11.9µg/L) and (11.3 ± 14

µg/L) (**Fig. 40**). The range of As concentration in urine samples of the male and female population was <3 to 48.3 µg/L (n=69) and<3 to 80 µg/L (n=66) (**Table 30**).

Table 30: Status of arsenic concentration in drinking water, rice grain and biomarkers of the studied population

Parameter	Drinking water As (µg/L)	Rice grain As (µg/kg)	Urine As (µg/L)		Hair As (mg/kg)		Nail As (mg/kg)	
Sex	-	-	**Male**	**Female**	**Male**	**Female**	**Male**	**Female**
N	50	28	69	66	58	60	58	60
Mean	176	211	10.1	11.6	4.14	4.33	8.41	7.69
SD	237	167	9.51	13.5	3.74	5.37	7.11	7.84
Median	41.3	143	6.65	7.34	3.38	2.16	6.32	4.78
Range	<3-847	52-668	<3-48.3	<3-80	0.04-17.6	0.04-24	0.3-28	0.35-29
Allowable or recommended limit*	10[a]	200[b]	3.33-26.7[c]		0.08-0.25[d]		0.43-1.08[e]	

*[a]WHO, (2011); [b] European Commission, (2015); [c] Farmer and Johnson (1990); [d] Arnold et al., (1990); [e] Ioanid et al., (1961)

Estimation of accumulated As in hair and nail is the best possiblemethod to apprehend long-term As exposure, followed by dermatological expressions (Shankar et al., 2014; Wade et al., 2015). Arsenic toxicityinduced effects on human health arises after 60 to 150days for hair samples and 12 to 18 months for nails, as conveyed by Nowak and Kozlowski (1998) and Pororinskaya and Karpenko (2009). Hair As concentrations were 0.84 ± 0.76 mg/kg (range: 0.38-2.7 mg/kg) in children, 3.07 ± 1.73 mg/kg (range: 0.64-5.73 mg/kg) in teenagers and 4.41 ± 4.31 mg/kg (range: 0.03-24 mg/kg) in adults in the present study, whereas, concentrations of nail As were 2.38 ± 1.62 mg/kg (range: 0.5-5 mg/kg), 6.18 ± 4.73 mg/kg (range: 0.55-14.8 mg/kg) and 9.07 ± 6.48 mg/kg (range: 1.14-25.3 mg/kg) in children, teenagers and adults (Fig. 40).

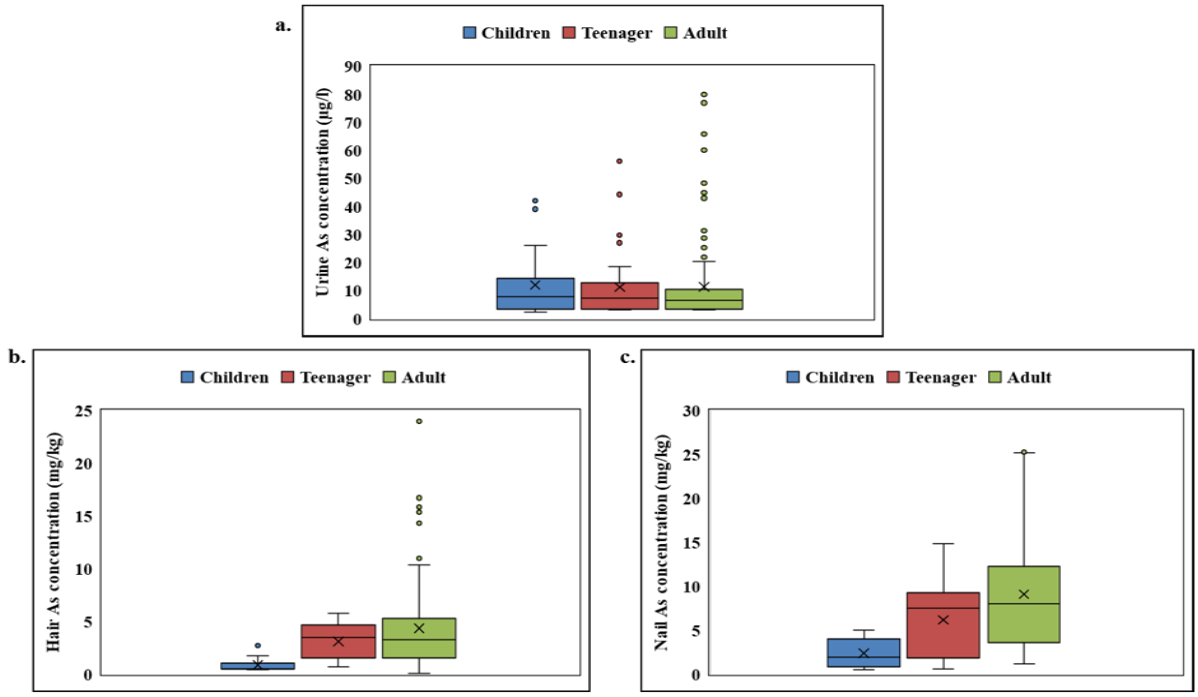
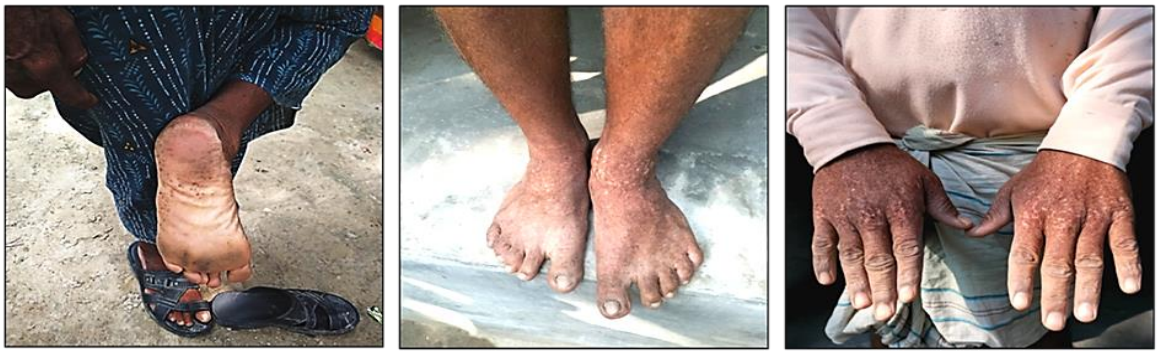


Fig. 40. Box plot showing accumulation of arsenic in biomarkers of the three differently aged population: (a)Urine (b) Hair (c) Nail

It is realized that deposition of As in hair and nail rises with age. It is augmented by 3.65 and 2.6 timesfrom children to teenagers and 1.44 and 1.47 times from teenagers to adults. Nail As deposition is higher than hair As deposition in children, teenagers and adults by 2.83, 2.01 and 2.05 times respectively. On the contrary, the mean nail As concentration of the male and female participants (8.41± 7.11 and 7.69 ± 7.84 mg/kg) was perceptiblygreater than that of hair (4.14 ± 3.74 and 4.33 ± 5.37 mg/kg) (Table 30). This stands with NRC (1999) that the degree of As accumulation is quicker in nail parts than hair. Higher mean nail As concentration than hair was also observed in other countries like Vietnam (Nguyen et al., 2019); northeastern Thailand (Wongsasuluk et al., 2018) and Cambodia (Gault et al., 2008).

5.4.2.5. Arsenic mediated dermatological expressions

The arsenic mediated body loadunveils that manyinhabitants are sub-clinically affected without any predominant skin lesion. Few individuals fro m the severely exposed population are identified with As persuaded symptoms. Arsenic caused dermal manifestations of affected individuals from the present study area are delineated in **Fig. 41**.

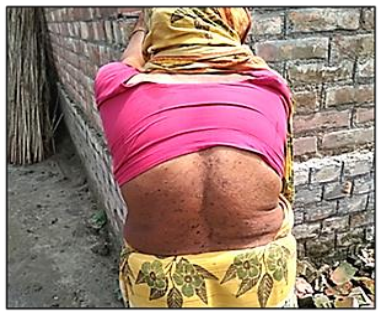


a. Keratosis on sole

b. Leucomelanosis, raindrop pigmentation

c. Raindrop pigmentation; Leucomelanosis

d. Spotted and diffused keratosis on palm



e. Spotted and diffused melanosis

f. Spotted and diffused keratosis on palm and sole; bowens

Fig. 41. Dermatological manifestations in affected individuals of the study area

Skin lesions are detected among the folks such as spotted and diffused keratosis, melanosis, and raindrop pigmentation in hand, feet or whole bodyeven Bowens. Also, Leuco-melanosis is found in a few sternly affected persons who have ceased drinking As-contaminated water but had spotted melanosis before (**Table 31**).

Table 31: Detailed informationof the chronic arsenic patients in the studied population

Case number	Patient Sex and Age	Location	Drinking water Arsenic conc. (µg/L)	Urine Arsenic conc. (µg/L)	Hair Arsenic conc. (mg/kg)	Nail Arsenic conc. (mg/kg)	Symptoms observed	Figure number
1	M, 50	Vill: Kadamtola; G.P: Katlamari- I	19.6 (tube-well)	NA*	5.04	15.34	Keratosis on sole	Fig. 41a
2	M, 48	Do	<3 (Sajaldhara treated water)	NA	0.94	1.26	Leucomelanosis, raindrop pigmentation	Fig. 41b
3	M, 50	Do	9.4 (pipeline)	4.92	2.28	6.85	Raindrop pigmentation; Leucomelanosis	Fig. 41c
4	F, 58	Do	408 (tube-well)	8.7	0.24	1.62	Spotted and diffused keratosis on palm	Fig. 41d
5	F, 62	Do	NA	NA	1.11	2.45	Spotted and diffused melanosis	Fig. 41e
6	M, 57	Do	310 (tube-well)	NA	3.88	4.80	Spotted and diffused keratosis on palm and sole; bowens	Fig. 41f

*NA: Not Available

Previously, researchers found that 19% of people with discrete arsenical skin manifestations among the 25,274 villagers; the most prevailing symptom was 'spotted and diffused melanosis on trunk' according to Mukherjee et al. (2005).

5.4.2.6. Arsenic exposure with reference to 'intake of arsenic'

A spearman's correlation ismade at 95% confidence level to evaluate the relation among ingested As and biomarkers As concentration (**Fig.42a**).

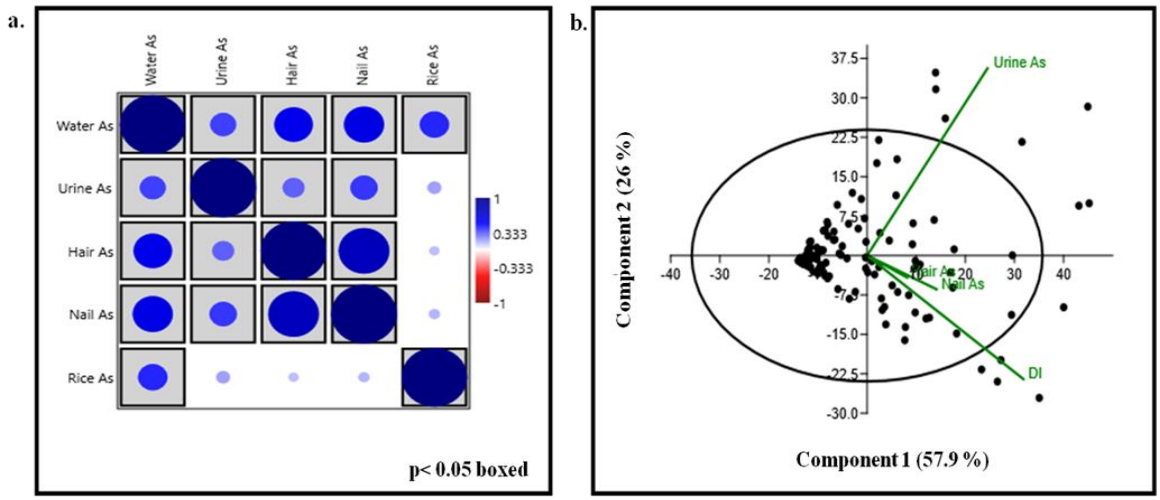


Fig. 42. Relation between arsenic intake and excrete: (a) Spearman correlation (b) Principal component analysis

Arsenic concentration in drinking water is observed to be correlated most strongly with nail samples (r = 0.60) compared to hair (r= 0.57) and urine samples (r = 0.38). Whereas, As concentration in rice grain is correlated poorly with urine (r = 0.18), hair (r = 0.13) and nail (r = 0.14). The correlation values (r) indicate that drinking water As accumulates in the keratin tissues slowly but strongly (**Table 32)**. As a result, deposition of As in the chronic biomarkers is an calmer way to estimate As toxicity in comparison to the acute biomarkers; stated in Brima et al. (2006) and Samanta et al. (2004).

Table 32: Spearman correlation among the intake and excrete variables

	Water As	Urine As	Hair As	Nail As	Rice As
Water As		0.37	0.57	0.60	0.43
Urine As	0.37		0.31	0.39	0.18
Hair As	0.57	0.31		0.76	0.13
Nail As	0.60	0.39	0.76		0.14
Rice As	0.43	0.18	0.13	0.14	

Arsenic in drinking water always affects more than other pathways as it is the direct way of i-As (Tokunaga et al., 2005; Rakhunde et al., 2012). A considerably decreased As concentration was observed reportedly in biological samples of few children residing in North 24 Parganas district, West Bengal, when they were provided with As-safe water (treated surface water) (Joardar et al., 2021b). In many As exposed regions like Thailand (Rujiralai et al., 2018) and Cambodia (Sthiannopkao et al., 2010), As-tainted drinking water is a verified exposure route in scalp hair samples. A momentous dependence was found between 'As intake/day through drinking water/body weight' and 'hair As concentration' in some As exposed populations from Nadia district, West Bengal. But, no significant interrelation was seen between 'As intake by diet' and 'hair As concentration' using a copula-based bivariate mixed regression (Das et al., 2018). Arsenic concentration in waterappeared to be the central route of exposure in toe-nail, reported formerlyin Signes-Pastor et al. (2021). Both hair and nail grow gradually and are alike in keratin structure, resulting into strong association between toenail and hair As concentration (**Fig. 42a**) which is substantiated in other reports (Signes-Pastor et al., 2021). Principal component analysis or PCA has been implemented next to comprehend the character of connection between DI (dietary As intake) and As concentration in the biomarkers (**Fig. 42b**). PCA is generally performed to reduce the dimensions of a data set by measuring correlation amongst the factors in form of principal components (De et al., 2022; Wenning and Erickson, 1994). This visibly explains that the vectors of DI, hair As and nail As are closely proximated to each other (quadrant 1) explaining that they are related to other directly. On the contrary, DI and urine As are placed in two opposite quadrants representing no direct association. The two major components demonstrated 47.5% and 33.9% variance (**Table 33**). It can therefore be concluded that the daily intake of As is manifested through the chronic biomarkers (hair and nail) directly in lieu of urine (Fig. 42b). The hair and nail tissues are keratin made (scleroprotein), enriched with cysteine remainders that contain sulfhydryl groups where As gets bounded easily (Gault et al., 2008). Therefore, the most constructive markers of metal exposure on human body systems are keratin, conveyed in many articles (Byrne et al., 2010; Samanta et al., 2004; Kumar et al., 2021).

Table 33: Principal component analysis among the variables (DI, Hair As, Nail As, Urine As)

Component	Eigen value	% Variance
1	205.56	57.947
2	92.4739	26.068
3	48.5038	13.673
4	8.19956	2.3114

A Kruskal Wallis test followed by a Dunn's post hoc test has validated the association between As intake and excretion through the two chronic biomarkers todiscover the most apt biomarker for evaluation of As toxicity. Kruskal-Wallis test is generally used to check the existence of any statistically significant difference between the medians of the nominated groups (**Table 34**).

Table 34: Hypothesis testing among DI and arsenic in chronic biomarkers through Kruskal- Wallis test followed by Dunn's post hoc test ($\alpha = 0.05$)

Hypothesis assumed:			
H_o	**No significant difference exists among the three groups (DI, Hair As, Nail As)**		
H_1	**Significant difference exists among the three groups (DI, Hair As, Nail As)**		
Kruskal-Wallis test for equal medians:			
H (χ2)= 21.05; H (crit) (Yates corrected)=21.05; p= 2.689×10^{-5}			
Remarks: There is a significant difference between two group sample medians (**H_o is rejected)**			
p value in Dunn's post hoc test:			
	DI (Daily intake of As) (n= 118)	**Hair As (n= 118)**	**Nail As (n= 118)**
DI		0.6877	0.0001721
Hair As*	0.6877		3.199×10^{-5}
Nail As	0.0001721	3.199×10^{-5}	

***$H_o$is rejected for Hair As (p > 0.05)**

'DI' is accounted as an independent factor and 'Concentrations of As in biomarkers' is dependent variable. A significant difference is found between sample medians at 95% confidence interval, as p value is less than 0.05 and the χ2 value is 21.05 (N = 354). Dunn's post hoc test is executed to know which particular group makes the difference. The null hypothesis is rejected for 'hair As' as it shows a non-significant p value towards daily intake of As (0.69), while 'nail As' is significant with a p value of 1.72×10^{-4}. Table 34 mentions the hypothesis assumed for the test along with the p values. Cumulatively, it can

be said that the nail samples are the most crucial biomarker for estimation of As toxicity. Gutiérrez-González et al. (2019) and Signes-Pastor et al. (2021) also reported that toe-nails are globally identified to have more steady and lasting metal depositions than the other biomarkers of acute exposures such as urine.

5.4.2.7. Arsenic exposure with reference to 'age'

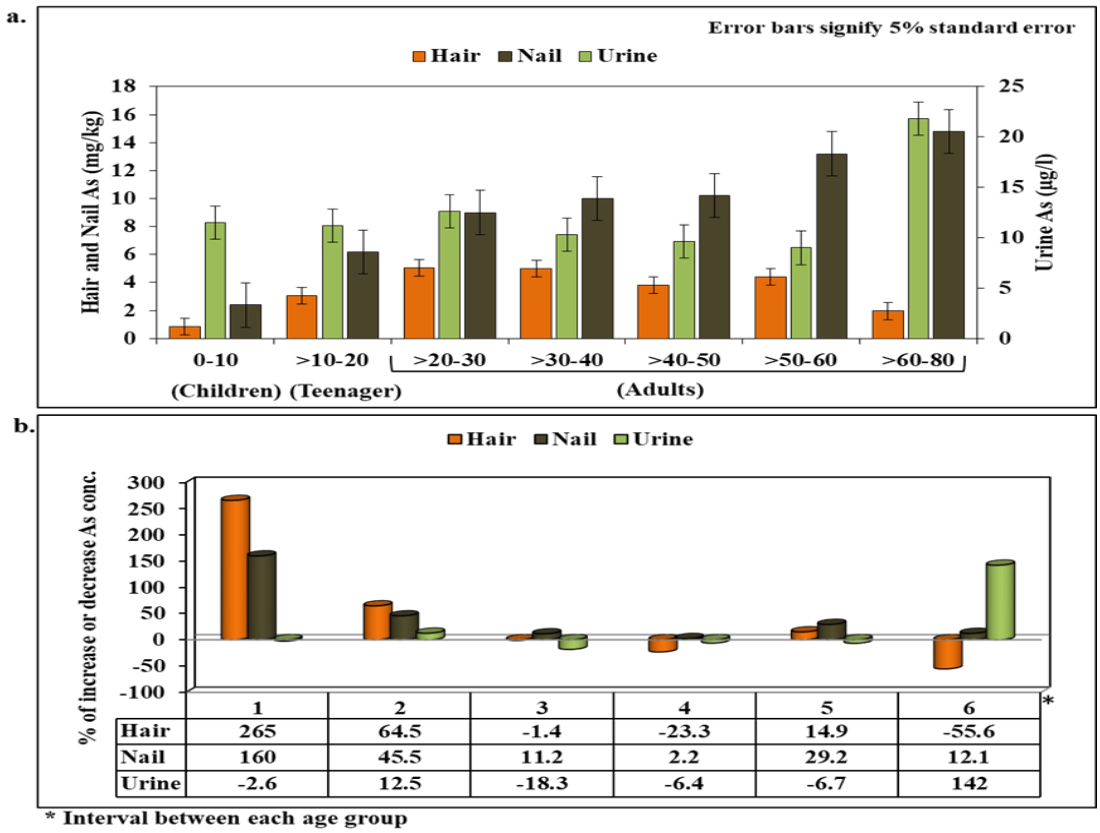
Arsenic accumulation in the biomarkers of the populace is appeared to be dependent on 'age'; explained through a 'two sample paired t test' (**Table 35**).

Table 35: Dependence of age on arsenic deposition in biomarkers of the studied population (two sample paired t test)

Hypothesis assumed:						
H_o	**No significant difference exists between age and As concentration in biomarkers**					
H_1	**Significant difference exists between age and As concentration in biomarkers**					
	Age (yr)	**Urine As (µg/L)**	**Age (yr)**	**Hair As (mg/kg)**	**Age (yr)**	**Nail As (mg/kg)**
Mean	33.4	11.1	34.4	4.02	34.4	9.16
Variance	259	124	257	20.4	257	74.2
Observations	127	127	116	116	116	116
Pearson Correlation	-0.00027		0.111		0.322	
Hypothesized Mean Difference	0		0		0	
Degree of freedom (df)	126		115		115	
t_{stat} one tail	12.83		20.2		17.4	
p (T<=t) one-tail	6.35×10^{-25}		6.11×10^{-40}		2.23×10^{-34}	
t_{crit} one-tail	1.657		1.658		1.658	
p (T<=t) two-tail	1.27×10^{-24}		1.22×10^{-39}		4.46×10^{-34}	
t_{crit}two tail	1.98		1.98		1.98	
Remarks	H_o is rejected		H_o is rejected		H_o is rejected	

A hypothesis is checked to confirm whether any significant difference is present between 'age' and 'As concentration in the biomarkers'. Null hypothesis (H_o) is rejected in each case, i.e. there exists a substantial difference between age and As concentration in the biological indices. The t_{stat}values (12.83, 20.2 and 17.4) are found to be much higher than t_{crit}value (1.98); df = 126, 115 and 115 for urine, hair and nail respectively. A negative and moderate positive correlation is also observed between age and As concentration in urine (r = -0.00027), hair (r = 0.111) and nail (r = 0.322). The existing study is significant with reference to the trend of As deposition in the biomaterials per the age of the participants.

The deposition of As changes with age which indicates accumulation of metals occur with frequency and length of exposure that also dependent on occupation of work (Nathet al., 2008). Mean nail As concentration noticeably increases with increasing age (**Fig. 43a**); however, the increase %drops slowly (**Fig. 43b**).



	1	2	3	4	5	6
Hair	265	64.5	-1.4	-23.3	14.9	-55.6
Nail	160	45.5	11.2	2.2	29.2	12.1
Urine	-2.6	12.5	-18.3	-6.4	-6.7	142

Fig. 43. Arsenic exposure with age: (a) Deposition of arsenic in biomarkers in seven subsequent age groups (b) Percentage variation of biomarkers' arsenic concentration between each age group

The mean nail As concentration is maximum in the old aged people aged (60 to 80 years) (14.8 mg/kg) and least in children aged below 10 years (2.38 mg/kg). Hair and nail As concentrations rise highest in teenagers from children by ~ 265 and 160% while it shrinkages towards the adults. Concomitantly, urinary As does not follow any proper pattern. Deposition of As drops slowly in the three subsequent age groups starting from 30 - 60 years by 18.3, 6.4, and 6.7% and surges abruptly in the old age group by 142% (Fig. 43b). Mean As concentrations in urine and hair are found to be maximum in participants of >60 to 80 years (21.9 µg/L and 14.8 mg/kg), while the mean hair As concentration (1.95

mg/kg) is relatively less than other adult groups (Fig. 43a). About 50 to 70% of engrossed As is eradicated in kidneys by methylation and the remaining As is deposited in hair, nail and other tissues; stated by Nielsen (2001). Arsenic concentration in nail is elevated than hair As in each age gap by roughly 3.13 times that might be because of higher As binding proteins in nail compared to hair accompanied by a slower growth rate (Mandal and Suzuki, 2002; Maity et al., 2012; Tobin, 2005). Thus, nail As concentration can be called as the best efficient biomarker for As toxicity regardless of a person's age. Deposition of As in body tissues occur in accordance to individual's age and the forte of metabolism (Mitra et al., 2004). In North 24 Parganas, another worst As affected part of Bengal, Maity et al. (2012) found that As concentration in the biomarkers of few population escalates with age and drops after 57 years. Bibi et al. (2015) presented that the distribution of As in Pakistan is maximum in adults followed by children and old people and it is concentrated in nail and blood samples followed by urine and hair. Fang et al. (2019) reported that the inhabitants from Huainan city (China) observed a rising hair As concentration with age. However, Rasheed et al. (2019) observed no significant consequence of age on deposition of As in chronic biomarkers.

5.4.2.8. Arsenic exposure with reference to 'gender'

In the physical and socio-economic well-being of As exposed people in the Murshidabad district, 'sex' was found to be a dominant influencer (Das, 2013). The mean As concentrations in urine (11.6 ± 13.5 µg/L) and hair (4.33 ± 5.37 mg/kg) in the female candidates is roughly 1.15 and 1.05 times bigger than the male candidates (10.1 ± 9.51 µg/L and 4.14 ± 3.74 mg/kg). Although, the mean nail As concentration in male population (8.41 ± 7.11 mg/kg) is ~ 1.09 times higher than the female population (7.69 ± 7.84 mg/kg). Exposure of As with reference to gender is tested through the deposition of As in the biomatrix of the studied populace. A $\chi 2$ test is conducted where the null hypothesis (Ho) is considered as 'As concentration in biomarkers does not depend on gender'. As concentration in the biomarkers of the male and the female populace is grouped in accordance to their individualstandard range (**Table 36**).

Table 36: Dependence of gender on arsenic deposition in biomarkers of the studied population ($\chi 2$ test; Yates correction included)

Hypothesis assumed:									
H_o	**As concentration in biomarkers is independent of gender**								
H_1	**As concentration in biomarkers depends on gender**								
Biomarkers	**Urine**			**Hair**			**Nail**		
Category or Group	**As conc. > Normal range**	**As conc. < Normal range**	**Total count (n)**	**As conc. > Normal range**	**As conc. < Normal range**	**Total count (n)**	**As conc. > Normal range**	**As conc. < Normal range**	**Total count (n)**
Male	5 (7.25%)	64	69	56 (96.6%)	2	58	53 (91.4%)	5	58
Female	7 (10.5%)	60	67	55 (91.7%)	5	60	55 (91.7%)	5	60
Degree of freedom (df)	1			1			1		
$\chi 2_{cal}$	0.13			0.54			0.075		
p value	0.72			0.46			0.78		
$\chi 2_{crit}$	3.84			3.84			3.84		
Remarks	$H_o$is accepted			H_o is accepted			H_o is accepted		

Near about 7.25% male participants (n=69) have higher As concentration in urine than the acceptable range while the female percentage is 10.5 (n=67). Higher toxic metals in urine were witnessed in women than men that probably caused by their increased gastrointestinal absorption at micronutrient deficiencies (Berglund et al., 2011). Noticeably, the extent of chronic As exposure in male and female populaces is too high compared to the acute exposure. The existing study found 96.6% male and 91.7% female hair samples with As concentrations beyond the allowance. While, in 91.4% of male and 91.7% of female populace (n=58 and 60), nail As is found to be higher than the normal range. Rahman et al., (2005a) found 95%, 75% and 94% of the nail, hair and urine samples with As concentration more than the normal levels among total of 3800 biological samples of the affected villagers. Also, Rahman et al. (2005b) found that from the affected villages of Sagarpara gram panchayats, Murshidabad, 85% of biological samples (n= 850) to contain As above normal range. The calculated $\chi 2$ value (0.13, 0.54 and 0.075) is found to be less than the critical value (3.84) for urine, hair and nail resulting into acceptance of the Howhich means that As deposition is not regulated by the sex of the mass. Similar observations were found by Das et al. (2018) where gender was neither an important forecaster of hair As concentration nor of any dermal manifestations. Though, few researches proved that sex plays an imperative role in As metabolism. Rahman et al. (2006) reported that the male population in Bangladesh are more susceptible to have As mediated

skin lesions than the female population when get exposed to As-contaminated tube-well water. Tseng (2009) submitted that adult women have an amended methylation ability of As compared to men, which can be explicated by the encouraging effect of estrogens on the amalgamation of choline, involved in the re-methylation of homocysteine. Pregnant women had better As methylation efficacy than men due to the sex hormones (Lindberg et al., 2008).

5.4.2.9. Assessment of health risk

A health risk evaluation has been done (eq. 9 to 11) on the studied population to assess the magnitude of upcoming intimidations caused by continued As exposure. **Fig. 44** along with **Table 37** explain that the substitute drinking water sources deliver adequate health risk as well.

Table 37: Cancer and non-cancer risk assessment for studied three age groups through different dietary sources

Target population		ADD		HQ		CR	
		Mean	Range	Mean	Range	Mean	Range
Children	Tube-well water (n=8)	3×10^{-3}	9.72×10^{-6} – 1.37×10^{-2}	10.08	0.032-45.7	4.53×10^{-3}	1.5×10^{-5}-2×10^{-2}
	Sajaldhara plant treated water (n=8)	9.95×10^{-4}	3.32×10^{-5}- 6×10^{-3}	3.32	0.11-20.11	1.49×10^{-3}	5×10^{-5}- 9×10^{-3}
	Pipeline water (n=2)	3.24×10^{-4}	1.07×10^{-4}-5.43×10^{-4}	0.90	3.19×10^{-8}-1.81	4.43×10^{-4}	7.10×10^{-5}-8.14×10^{-4}
	Rice grain (n=8)	3.3×10^{-4}	1.19×10^{-4}- 8.43×10^{-4}	1.10	0.4-2.81	4.95×10^{-4}	1.8×10^{-4}-1.27×10^{-3}
Teenager	Tube-well water (n=15)	5.39×10^{-3}	1.07×10^{-4}-1.19×10^{-2}	17.99	0.36-39.9	8.1×10^{-3}	1.6×10^{-4}-1.79×10^{-2}
	Sajaldhara plant treated water (n=7)	2.69×10^{-3}	1.1×10^{-4}-6.8×10^{-3}	8.97	0.37-22.7	4.04×10^{-3}	1.7×10^{-4}-1.02×10^{-2}
	Pipeline water (n=4)	3.2×10^{-3}	1.63×10^{-4}-7.3×10^{-3}	1.33	0.55-2.44	6×10^{-4}	2.5×10^{-4}-1.09×10^{-3}
	Rice grain (n=18)	4.12×10^{-4}	1.29×10^{-4}-1.18×10^{-3}	1.38	0.43-3.96	6.19×10^{-4}	1.9×10^{-4}-1.78×10^{-3}
Adult	Tube-well water (n=92)	7.68×10^{-3}	8.52×10^{-5}-5.2×10^{-2}	34.09	0.28-186	1.53×10^{-2}	1.3×10^{-4}-8×10^{-2}
	Sajaldhara plant treated water (n=23)	4.55×10^{-3}	1.16×10^{-4}-1.64×10^{-2}	14.9	0.39-54.9	6.7×10^{-3}	1.7×10^{-4}-2.47×10^{-2}
	Pipeline water (n=6)	1.11×10^{-3}	4.45×10^{-4}-1.78×10^{-3}	4.65	1.48-8.59	2.09×10^{-3}	6.7×10^{-4}-3.87×10^{-3}
	Rice grain (n=53)	9.99×10^{-4}	1.23×10^{-4}- 3.13×10^{-3}	3.72	0.4112.5	1.67×10^{-3}	1.85×10^{-4}- 5.6×10^{-3}

The children who ingested water from tube-well, suffer from higher risk of cancer (range: 1.5×10-$5^{-2}\times10^{-2}$) than those who drink alternative water (Fig. 44). Water from Sajaldhara

water treatment plants and pipeline systems, both being the substitute drinking water sources in the As exposed zone, impose high risk of cancer in children (range: 5×10^{-5}-9×10^{-3} and 7.10×10^{-5} - 8.14×10^{-4}) than the acceptable value. Mean cancer risk from tube-well water is found to be 8.1×10^{-3} and 1.53×10^{-2} for teenagers and adults, respectively (Table 37). The respective mean cancer risk through treated water from Sajaldhara plants is calculated as 4.04×10^{-3} for teenager and 6.7×10^{-3} for adults. Arsenic concentration in rice grain is also a promising agent to cause cancer risk as the estimated average values are 4.95×10-4 (range: 1.8×10^{-4}-1.27×10^{-3}), 6.19×10-4 (range: 1.9×10^{-4}-1.78×10^{-3}) and1.67×10^{-3} (range: 1.85×10^{-4}-5.6×10^{-3}) for the three classes of populaces. The calculated values reveal that the population faces a substantial health risk that might cause cancerous diseases in future. The studied age groups have a considerable non-cancer risk through each source of drinking water in addition to rice grain in comparison to the USEPA onset value. The estimated mean non-cancerous risk is high in adults than teenager and children; HQ (tube-well water) values are 34, 18 and 10; HQ (Sajaldhara plant treated water) values are 14.9, 8.97, and 3.32; and HQ (pipeline supplied water) values are 4.65, 1.33, and 0.90 for adults, teenagers and children. Lowest amount of HQ is provided by contaminated rice grain, sufficient to cause non-cancer risk (mean values are 1.10 for children; 1.38 for teenagers; 3.72 for adults) (Fig. 44). Thereforeit can be said that the health risk follows: tube-well water >Sajaldhara treated water> rice grain > pipeline supply for children and teenagers while for adults: tube-well water >Sajaldhara treated water> pipeline supplied water > rice grain.

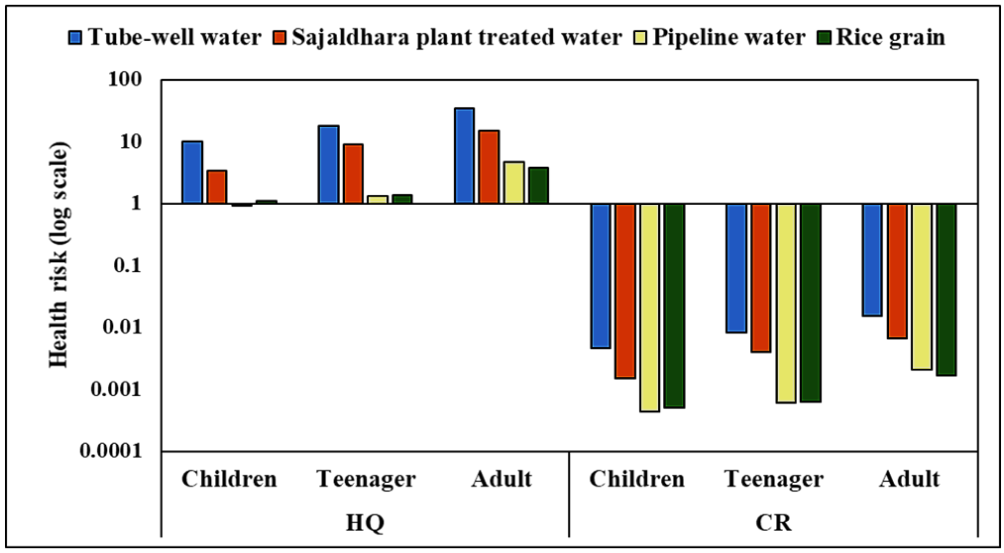


Fig. 44. Health risk assessment of the three studied age groups through different dietary sources

Results state that health risk of a population depends on several issues like metal concentration in source, exposure duration of the participants (age), body weight etc. Malnutrition in rural Bengal is also one more likely cause to suffer from from As toxicity. The dug wells are reportedly the anodyne source of drinking water in the studied area. They observed <3 to 5.5 µg/LAs concentration in the dug well water samples with an average of 3.78 µg/L. So, the authors suggest to increase the usage of dug-wells and surface water for drinking as well as cultivation purposes.

5.4.2.10. Relative importance of the variables used in assessment of health risk

5.4.2.10.1. Monte Carlo (MC) simulation along with sensitivity analysis

During the assessment of health risk, high uncertainty is observed when single fact values are used to evaluate the risk of a certain populace. MC simulation technique is used to enumerate the uncertainties of As exposure caused by drinking water and rice. In the simulation, a range of the variable is used repeatedly to achieve a degree of assurance in lieu of a single point value. The comparative importance of the factors related to the health risk of the populaces is evaluated through a sensitivity analysis, is shown through **Fig. 45**. It shows the input % of the variables (ED, IR, As C and BW) on the calculated health risk. A sensitivity analysis study is performed with HQ and CR values that reveals that As concentration (C) is the main factor operating in health risk amongst all the other variables. The impact of As concentration in drinking water to the estimated health risk is observed to be maximum in teenagers followed by adults and children (61.2%, 49% & 27.6%), while, it is maximum in adults (47.8%), followed by teenagers (45.5%) and children (21%) for rice grain. The 2ndand 3rd most significant factor in health risk of children is BW and ED; 47.5% and 8.97% by rice grain and 27.4% and 17.8% by drinking water, (Fig. 45). In adults, contribution of ED (17.2% and 20.1%) is higher than BW (9.26% and 12.1%) from both drinking water and rice grain; though in teenagers, the contribution of ED (4.62% and 5.81%) is lesser than BW (13.7% and 17.3%). For drinking water mediated risk, IR is the 4thimportant parameter with contribution % 1.36, 9.49 and 9.23 correspondingly in children, teenagers and adults. The input % of IR through As-contaminated rice grain are 5.63, 20 and 7.34 in the age groups (Fig. 45).

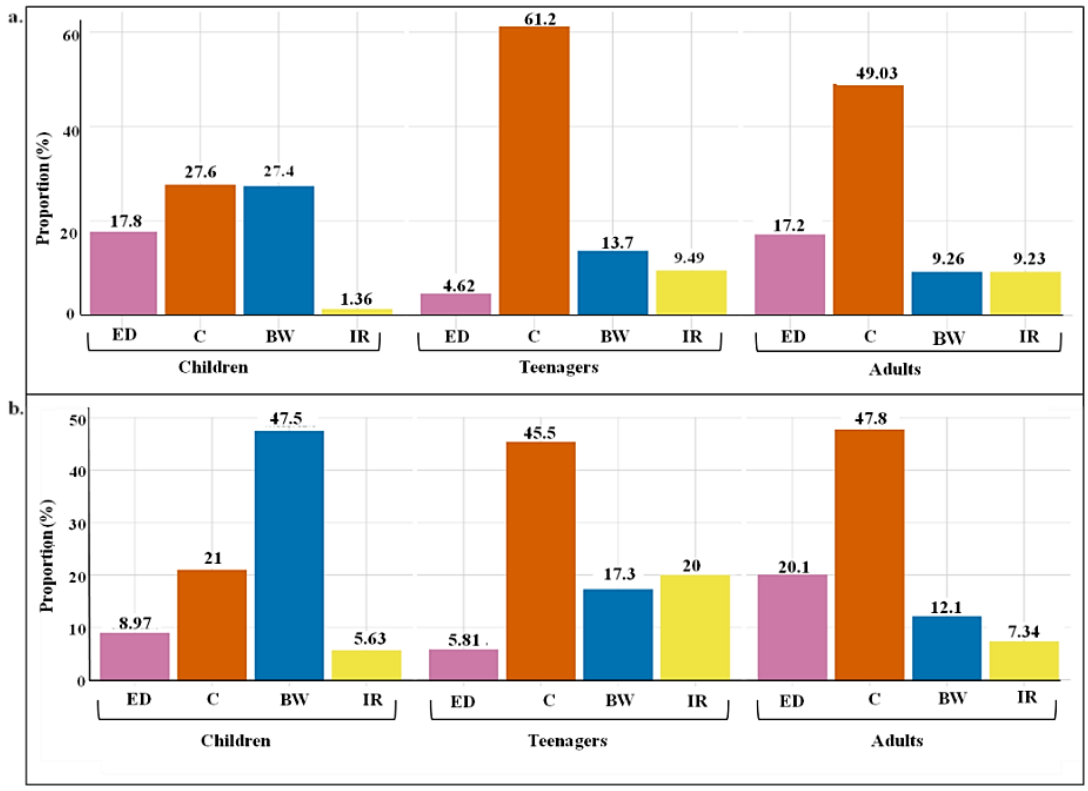


Fig. 45. Monte Carlo sensitivity analysis for health riskassessmentin the studied population: (a) drinking water (b) rice grain

Hence, it is decided that it is required to lessen the contamination at source to reduce the health risk. As consumption of As-contaminated rice grain also gives enough health risk, farming with higher As-contaminated irrigational water should be forbidden. Giri et al. (2020) supports these findings where they showed that the two most important contribution factors in assessment of the non-cancer risk of populaces are concentration of the metals in groundwater and exposure duration. Effect of Cwas (52.4 to 53.3) % and ED was (40.7 to 41.4) %, respectively in three age groups. Sharafi et al. (2019) and Pirsaheb et al. (2021) presented that 'metal concentration' and 'body weight' are the imperious aspects affecting the HQ value in few Iranian populations.

5.4.2.10.2. Hierarchical cluster analysis

A cluster analysis clusters the variables in different sized clusters according to their similar pattern through a dendrogram plot. **Fig. 46** indicates that metal concentration (C) is the main feature, directly connected to cancer and non-cancer risk (CR &HQ). CR and HQ are so narrowly arranged that their cluster is merged as they are analogous in nature. The discrete heights between 'C' and 'CR' or 'HQ' discloses that As concentration in drinking

water is more strongly allied to health risk than As in rice grain (Fig. 46a, b). The tiniest cluster prepared with intake rate and body weight (IR &BW) suggests that these depend on each other resulting into another association with exposure duration (ED).

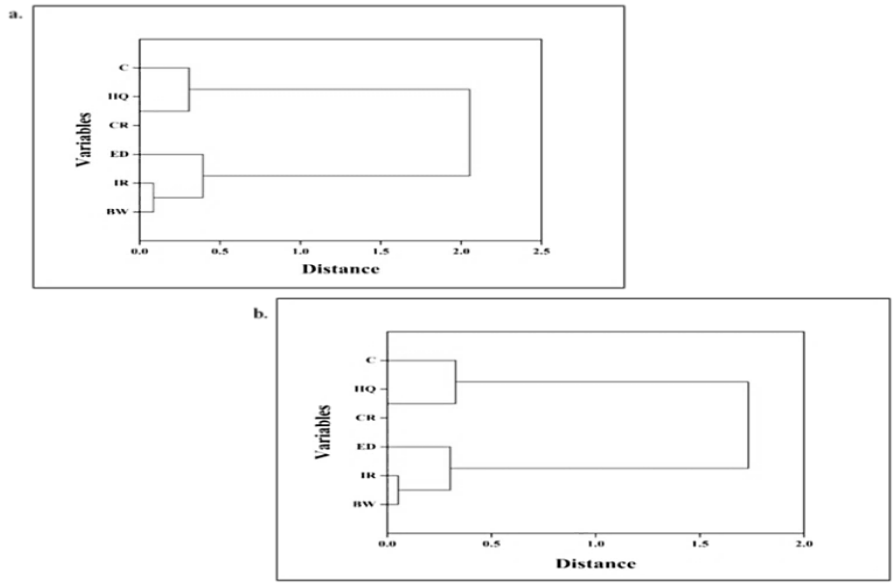


Fig. 46. Cluster analysis among the variables used in assessment of health risk through arsenic contaminated dietary items (a) Drinking water (b) Rice grain

5.5. Effect of arsenic toxicity on domestic livestock

Exposure of the geochemical As in domestic livestock with reference to cattle and goats through their biomarkers is the aim of the existing study. The daily intake of As and bio-concentration factor highlights the probability of severe toxicity. Arsenic assimilation patterns in fractions of whole milk that secreted from milch cows is also assessed in this study. The milk-products got from the local sweet-shops to apprehend the added health risk. Further evaluation is done on the reliance of excreted As on the body-system with different consumption pathways. It emphases on As exposure and human health risk caused by the most common daily diet comprising water, rice grain, and comestible animal goods through USEPA-based health risk assessment model and the application of 'risk thermometer'. The study searches the ecological risk through the 'bio-transformation factor' initiated by the domestic livestock that has the probability of depreciating the sustainability of the ecology.

5.5.1. Arsenic accumulation status in drinking water and fodder

Gaighata, a community development block under North 24 Parganas, is reportedly identified for extensive misery from As contagion over drinking water and foods (Roychowdhury, 2010). **Table 38** shows the present grade of drinking water and fodder As contamination from the study area. The range of As concentrations in drinking water of livestock from exposed sites are 4.8–403 µg/L (mean: 86.7 ± 99 µg/L) and 18–604 µg/L (mean: 309 ± 273 µg/L), respectively, which surpass the highest level of As in drinking water for heifers, (50 µg/L, NRC, 2001), while the As concentration from control sites is below 3 µg/L. Most of the families feed the livestock field crops and crop remainders. The As concentration in rice straw (433–5726 µg/kg) and husk (384–912 µg/kg) shows the extent of pollution in the agronomic wastes that are used as animal forage (Table 38).

Table 38: Arsenic accumulation status in drinking water, fodder and biomarkers of the studied livestock

Study area	Statistical Parameter	Drinking water As (µg/L)	Foodstuffs (µg/kg)									Excreta (Biomarkers)		
			Paddy Straw	Mustard cake	Corn Husk	Rice Husk	Crushed Rice Grain	Chickpea chaff	Maize	Wheat Chaff	Mixed cereal and husk	Urine (µg/L)	Faeces (µg/kg)	Tail Hair (µg/kg)
Exposed (Cattle)	N	30	30	30	25	28	30	22	22	26	-	30	30	30
	Mean	86.7	1940	94.8	178	591	429	117	345	139	-	9.53	794	698
	SD	99	1327	59.7	132	193	103	109	258	69	-	7.37	425	356
	Range	4.8-403	433-5726	22.5-203	19.8-790	384-912	197-779	21.2-330	155-712	57.4-228	-	<3-28.2	237-1804	175-1491
Exposed (Goats)	N	15	15	-	-	-	-	-	-	-	14	15	-	-
	Mean	309	2451	-	-	-	-	-	-	-	656	205	-	-
	SD	273	1536	-	-	-	-	-	-	-	467	108	-	-
	Range	18-604	450-4050	-	-	-	-	-	-	-	387-1319	92-338	-	-
Control (Cattle)	N	10	10	-	-	10	-	-	-	-	10	10	10	10
	Mean	3	543	-	-	182	-	-	-	-	100	3	243	254
	SD	0.78	271	-	-	56	-	-	-	-	71	2.07	53	209
	Range	<3-3.2	210-867	-	-	114-250	-	-	-	-	45-205	<3-7	184-319	75-502

It is relevant to state that the goats are frequently pastured in the local fields and their drinking water and feed sources differ. The feedstuff shows the As accrual arrangement in an order of paddy straw> rice husk> crushed rice grain> maize> corn husk> wheat chaff>chickpea chaff> mustard cake, which backs the downward As translocation pattern in paddy plant systems. As -contaminated groundwater produced rice, vegetables and othercereal crops fund a huge amount of i-As (Cubadda et al., 2017; Joardar et al., 2021a). Pal et al. (2007) reported upper range of As (2084–8394 µg/kg) in rice hay from Deganga block, North 24 Parganas. Mustard cake is made from the waste ingredients during the production of mustard oil, and mustard kernels contains50 ± 5 µg/kg of total As, where contribution of As-III is 48 ± 5 µg/kg, reported in Signes-Pastor et al. (2008). The silage is grown in an aboriginal As-contaminated soil, that coerces the domestic livestock to have a substantial i-As through their daily regimes. Paddy soil showed a mean As concentration of 37, 411 ± 10,281 µg/kg in Deganga (Chowdhury et al., 2018a). Therefore, soil subsidizes a raised-up level of As to the products grown in As-contaminated water. The feed containing paddy straw, husk and other cereals obtainable from the control sites displays radically low As than the exposed sites (Table 38).

5.5.2. Food habit and arsenic intake per day

Cattle feed covers paddy hay, grass silage, grains, husks etc. Generally, an adult cow or bull intakes40 L of water per day and 5 kg of straw. Supplementary 1 kg mixed cereals or protein complements are sometimes given as per their growth trend. Farther, 8 kg green food is provided to an adult cow for production of 1 L milk. **Fig. 47** explainsconcentration

of As in drinking water and feed and the dietary intake of As per day. Results show that in exposed area, rice straw gives 1940 µg/kg of As (range: 433 to 5726 µg/kg). Considering this routine, an exposed area adult cow/bull may have 13438 µg of As per day. Per capita intake of As through rice straw and water in exposed cattle population is ~ 3.57 and 37 times greater than the control population (Fig. 47). Hence, the total daily dietetic intake of As is almost 4.56 epochs more in exposed cattle to control. An adult domestic goat of exposed area consumes 0.7 kg of paddy straw, 0.4 kg of other mixed cereals and 5.5 l of water (range: 4-7 L), per day. Table 38 shows that the daily ingestion of As of a goat in exposed area is 3678 µg/day (Fig.47). Yet, Bera et al. (2010) and Raikwar et al. (2008) suggested the safe intake level of As for livestock is 15,000-25,000 µg/day. According to NRC (2005) and Roy et al. (2013), for livestock, the highest tolerable dietary intake of As was 50,000 µg/kg for i-As and 100,000 µg/kg for organic As, respectively. The maximum As content in complete feed of animals set by the European Union is 2000 µg/kg containing 12% moisture (EFSA, 2009; Mandal, 2017). Livestock from the exposed area devour a considerable quantity of As compared to the safe intake levels.

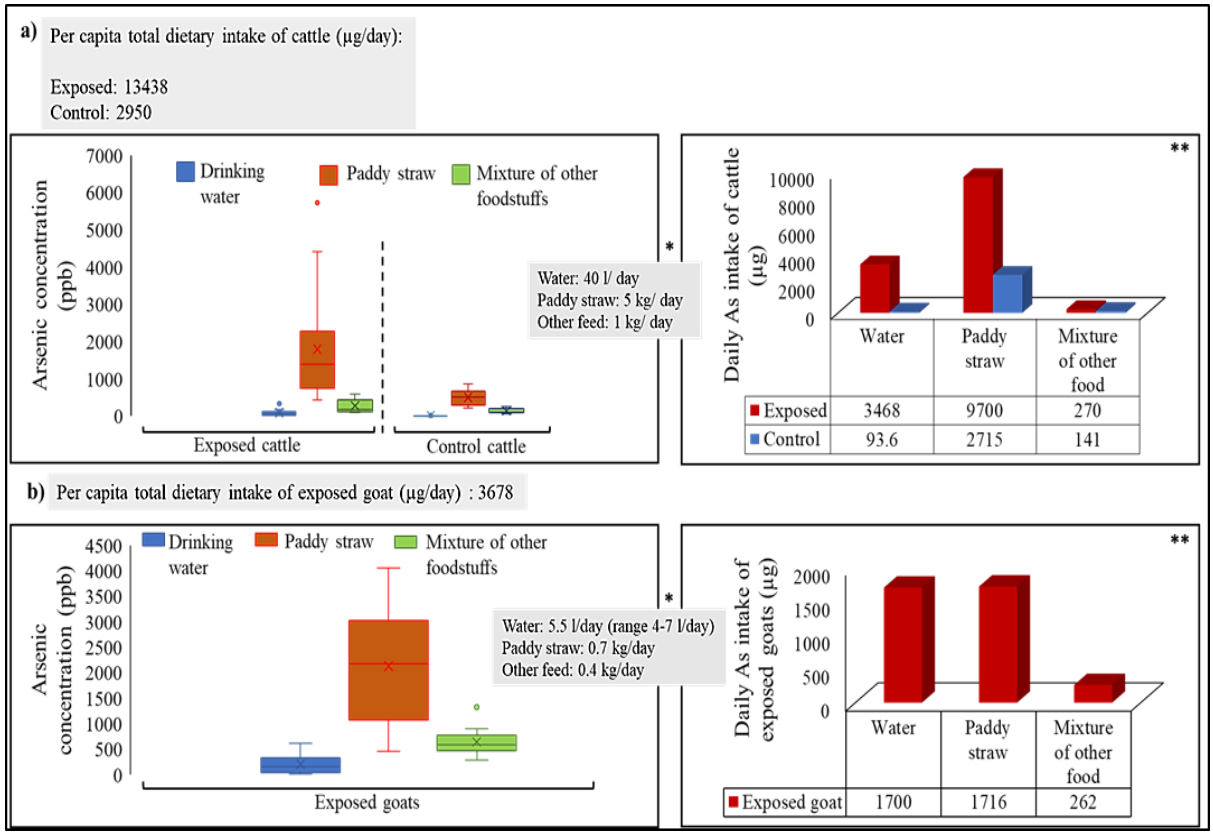


Fig. 47. Arsenic concentration in drinking water and fodder and daily dietary arsenic intake of domestic livestock

5.5.3. Accumulation of arsenic in biomarkers of the livestock

Urine and bile majorly express acute and sub-acute exposure, while faeces and hair evaluate chronic As exposure (Selby et al., 1974, 1977). The mean urinary As concentration (9.53 ± 7.37 µg/L) and dung As concentration (794 ± 425 µg/kg) of exposed cattle are greater than the control cattle population (3.0 ± 2.07 µg/L) and (243 ± 53 µg/kg), respectively. Urinary As concentration in exposed area cattle is within the tolerable limit of 50-170 µg/L (Lakso and Peoples, 1975). However, due to consumption of higher As through drinking water, the goats ensure high accrual of As in urine (mean: 205 ± 108 µg/L). The mean As concentration in tail hair of exposed cattle (698 ± 356 µg/kg), is even higher than the upper limit, i.e. 500 µg/kg suggested by Blood et al. (2000). Discharge of As is noticeable through faeces samples than urine which might be an outcome of small undigested part of dried food excluding pattern of livestock (Ghosh et al., 2013). Even if, a certain extent of trace element is not absorbed in body system as soil dirt imposes a definite exogenous exposure (Abrahams and Thornton, 1994). Due to the grazing exercise of livestock, the metals from soil powder get transported to the skin or hair of livestock in the dry seasons. Cattle do have a practice of licking their tails, or their calves' skins, which increases the oral consumption of pollutant, reported in Rogowska et al. (2009). Mandal (2017) reported that after ingestion of As, the concentration drops quickly in internal tissues, however transferred to the ectodermic tissues through sulfur containing proteins after several weeks. In most animal species, arsenical compounds go through controlled metabolism, enter the cell belatedly (ATSDR, 2007). Hair As gives the idea about the extent of contamination as hair is rich in keratin proteins that consists several sulphydryl confined amino acids (Hopps, 1977; Wilson and Lewis, 1927). Different foodstuffs contribute in the exposed population generously with As, while the relationship appears to be insignificant in the control subjects due to reduced contamination and possible improved methylation capability.

5.5.4. Role of drinking water arsenic on excreted arsenic

Regression analysis between water As and excreta gives the idea about the degree of As exposure from drinking water on livestock health (**Fig. 48**). Drinking water As and urinary As of cattle is correlated well from both the exposed (R^2 = 0.709, Fig. 48a) and control area (R^2 = 0.614, Fig. 48b), which further designates that most of the consumed i-As is excreted rapidly through urine. Similarly, the noteworthy correlation between As

concentration in water and faeces in exposed cattle population shows that secondary mode of elimination of toxic elements is surely defecation (Fig. 48a), however, in control populace it is less significant (Fig. 48b).Keratin cell expression is strong in the control population than exposed as the association between water As and tail hair Asseems stronger (R^2 = 0.752, Fig.49b) compared to the exposed populace (R^2 = 0.20, Fig.49a).In exposed goats, As exposure from drinking water is also revealed in urine through regression (R^2 = 0.435) (Fig. 48g).

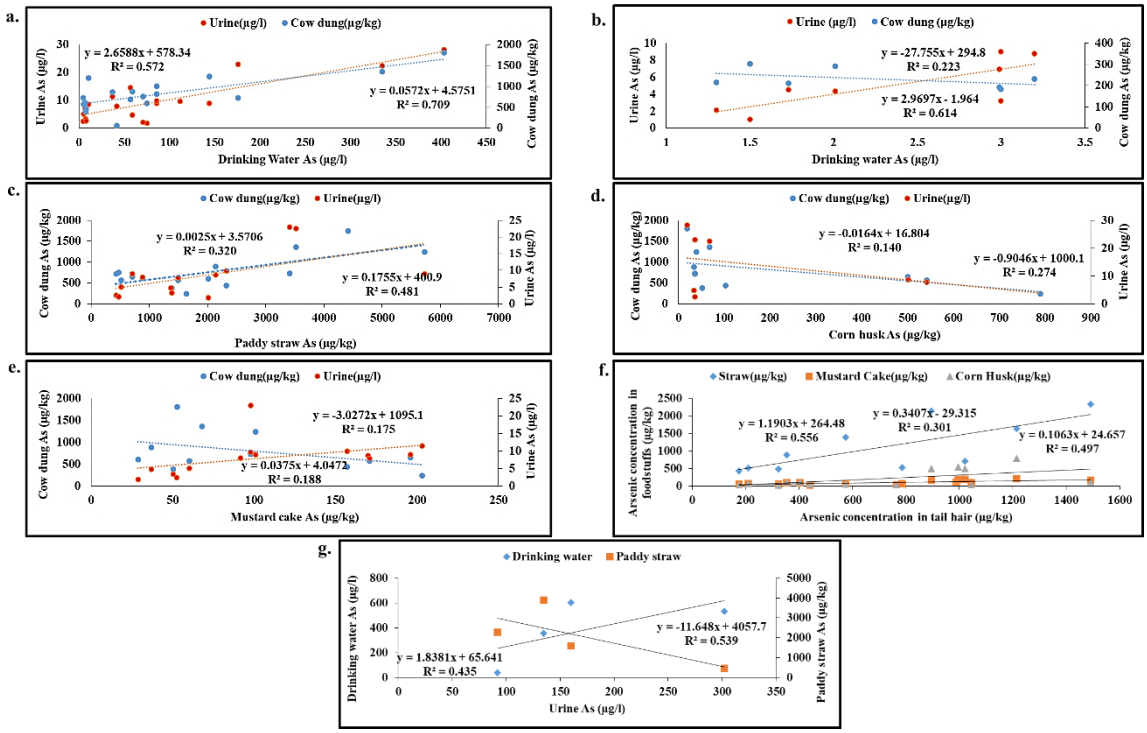


Fig. 48. Exposure of arsenic on livestock health through regression analysis: a) Relation between drinking water arsenic with urine and faeces arsenic in cattle (exposed area) b) Relation between drinking water arsenic with urine and faeces arsenic in cattle (control area) c) Relation between paddy straw arsenic with faeces and urine arsenic in cattle (exposed area) d) Relation between corn husk arsenic with faeces and urine arsenic in cattle (exposed area) e) Relation between mustard cake arsenic with faeces and urine arsenic in cattle (exposed area) f) Relation among arsenic in different foodstuffs and tail hair arsenic in cattle (exposed area) g) Relation between arsenic in drinking water and paddy straw with urine arsenic in goats (exposed area)

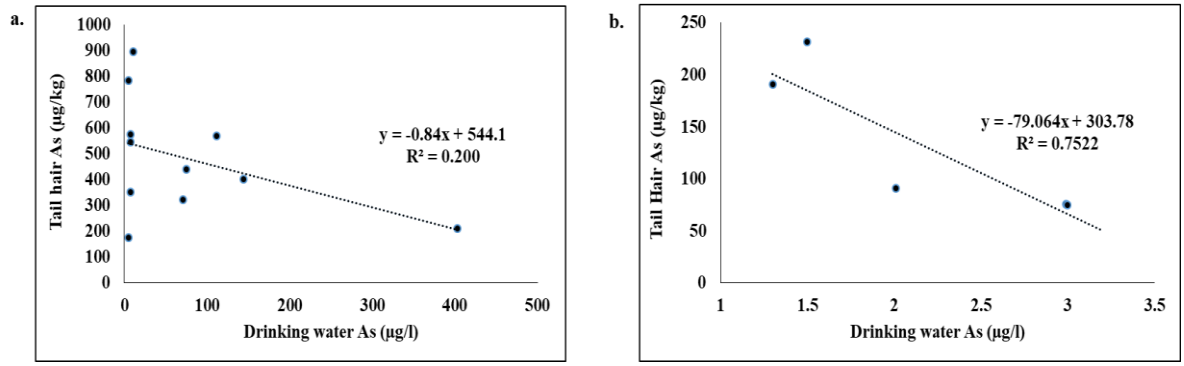


Fig. 49. Relation between drinking water and tail hair arsenic concentration of studied cattle population: a) exposed area b) control area

5.5.5. Role of food arsenic on excreted arsenic

Arsenic from paddy straw in exposed area is well excreted through urine (R^2 = 0.481) than faeces (Fig. 48c). Similarly, the influence of As from rice straw is articulated in exposed goats through urine (R^2 = 0.539, Fig. 48g). However, As from corn husk is expelled more in cattle dung than urine though the correlation values are not sturdy (R^2 = 0.274 and R^2= 0.14, respectively) (Fig. 48d). The accumulated As in mustard cake is excreted from body system through urine and faeces quite in the same way (R^2 = 0.188 and R^2= 0.175), the regression being weak (Fig. 48e). As concentration in tail hair also displays that rice straw, mustard cake and corn husk have substantial effect on sub-clinical toxicity (Fig. 48f). Rice straw and mustard cake As are expressed more in tail hair than corn husk, respective regression values are 0.301, 0.556 and 0.497. The effect of As from different harvests on excreta of the control cattle population is low as the extent of adulteration is very less. Rice straw has no noteworthy effect on urinary As (r = 0.096), faeces (r = 0.078) and awfully insignificant with tail hair (r = -0.62). Arsenic concentration in rice husk has a very trivial effect on tail hair (r = 0.123), but has neither any implication on urine (r = 0.087), nor on faeces (r = -0.152). Arsenic concentration in other grains also has less contribution on cattle dung (r = 0.31) while correlation between urine and tail hair As are non-significant (r = -0.088 and r = 0.032), respectively.

Table 39: Correlation matrix among arsenic in different foodstuffs and excreta in control cattle population

	Paddy straw (µg/kg)	Rice husk (µg/kg)	Food mix (µg/kg)	Urine (µg/L)	Cow dung (µg/kg)	Tail Hair (µg/kg)
Paddy straw (µg/kg)	1					
Rice husk(µg/kg)	NA	1				
Food mix (µg/kg)	NA	NA	1			
Urine (µg/L)	0.096 *	0.087 ×	-0.088 *	1		
Cow dung (µg/kg)	0.078 *	-0.152 **	0.310 **	NA	1	
Tail Hair (µg/kg)	-0.620 ***	0.123 **	0.032*	NA	NA	1

*p <0.05; **p <0.01; *** p≮ 0.05 or 0.01

5.5.6. A follow up study on accumulation of arsenic in biomarkers

A follow up study has been done on selected 4 cows for consecutive 3 months to examine whether the Astoxicity is varied with time or not. The range of As concentration in their drinking water and paddy straw samples were observed as 9.65-120 µg/L (average: 84.5 µg/L) and 563 – 1245 µg/kg (average: 845 µg/kg). Arsenic concentration in fodder and excreta samples was noted in **Table 40**.

Table 40: Arsenic concentration in fodder and excreta for consecutive 3 months

	Subject no.	Arsenic concentration (µg/L or µg/kg)				
		Drinking water	**Paddy straw**	**Cow dung**	**Urine**	**Tail hair**
Month 1	1	9.65	728	413	3.06	214
	2	120	845	506	5.56	258
	3	96.3	1245	1452	3.18	562
	4	112	563	942	5.56	1032
Month 2	1	9.88	728	542	3.04	326
	2	112	845	645	8.98	310
	3	115	1245	1789	3.54	458
	4	123	563	834	3	980
Month 3	1	8.45	728	456	3.21	312
	2	110	845	938	4.56	215
	3	95	1245	1208	4.9	780
	4	120	563	726	5.66	974

Month wise As accumulation in the biomarkers of the selected subjects has been shown in **Fig. 50**. Average As concentrations in cow urine samples were observed as 4.34, 4.64 and 4.58 µg/L in the three respective months while that in cow dung samples and tail hair samples were 828, 952, 832 µg/kg and 516, 518, 570 µg/kg. Accumulation of Asin excreta does not depend on time. It exclusively depends on the Asintake per day. Higher As intake leads to higher As bioaccumulation in body. The association between drinking water and cow dung, drinking water and tail hair were observed as R^2 = 0.60 and 0.53 whereas that between paddy straw and cow dung, paddy straw and tail hair were observed as R^2 = 0.83 and 0.13.

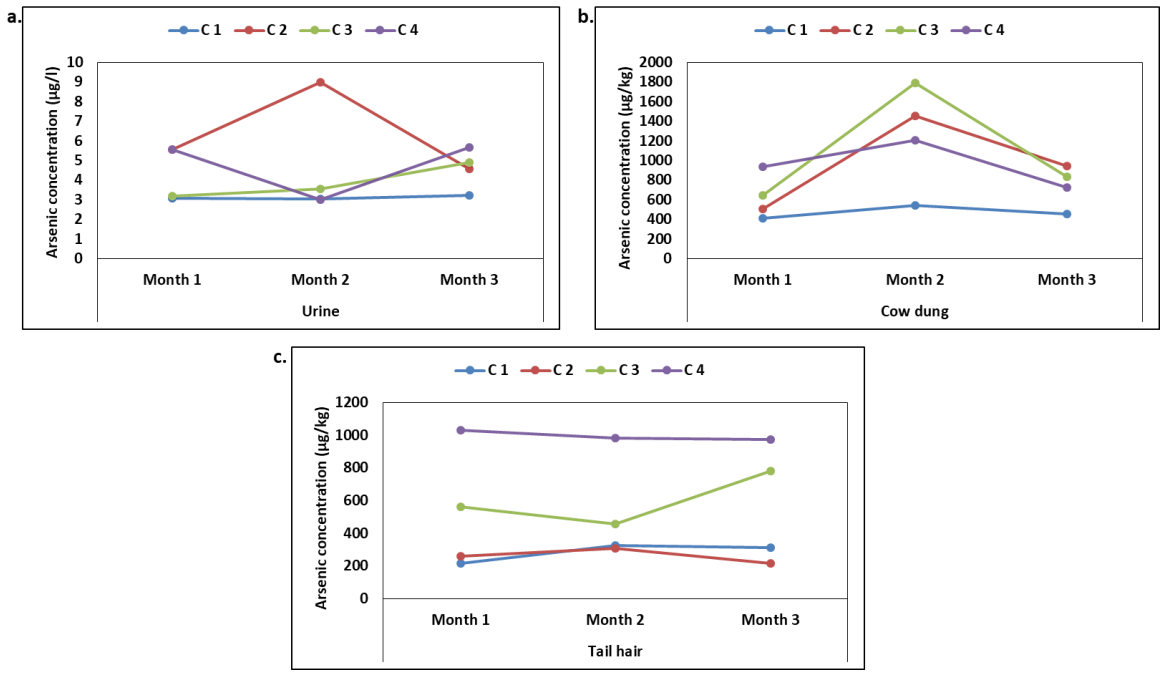


Fig. 50. Month-wise arsenic accumulation in biomarkers (C denotes number of cow)

5.5.7. Analysis of bio-concentration factor (BCF)

Accumulation of As through oral pathway is evaluated by Bio-concentration factor (BCF) (Liao et al., 2003). Rana et al. (2012, 2014a) said that it also signifies the degree of assimilation of the metal in body-system compared to the background level.

$$\text{Bioconcentration factor} = \frac{\text{As concentration in biological samples}}{\text{As concentration consumed orally (direct or indirect)}} \qquad \text{eq. (17)}$$

BCF calculation has been attempted by the quotient of Asconcentration in each excreta sample cattle and their oral ingestingpathways (**Fig. 51**). Thissubstantiates with the outcomes of Rana et al. (2012) showing bio-concentration procedure of As is faster through water in comparison to rice straw. In exposed cattle population, BCF is mostly

established in dung, followed by tail hair, while the course is tail hair followed by dung in control subjects. BCF_{milk} is least for both type of populations (Fig. 51a). The variation of BCF values through different pathways are an output of the digestive schemes of the heifers. Liver and kidney the prime organs where the xenobiotics are stowed and later join inbio-methylation (Roy et al. 2013). Datta et al. 2012) reported that accrual of As in tissue is slow, it is extreme in tail hair. It is to follow that BCF of As is larger in control cattle through every course than the exposed ones, perhaps due to the better metabolic activity.

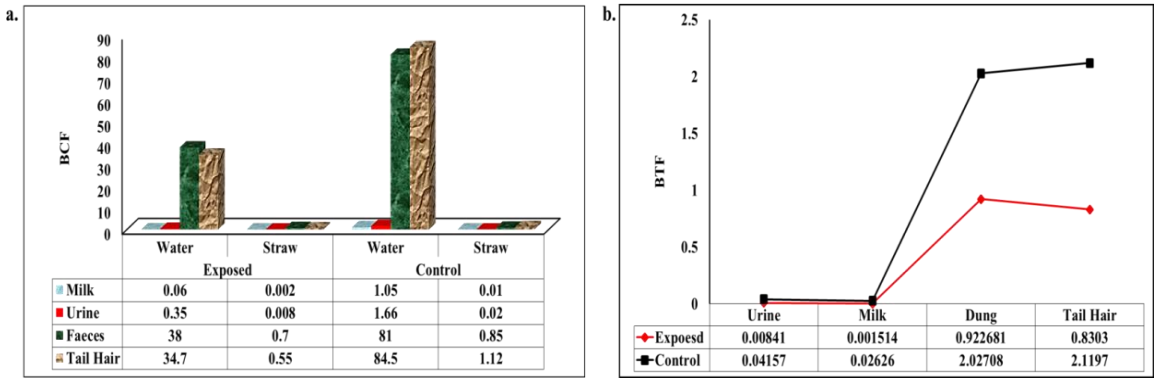


Fig. 51. Bio-concentration-and Biotransformation factor for livestock (cattle)

5.5.8. Arsenic concentration in animal proteins

Concentration of total As in frequently ingested animal-source foods (cow milk, meat, liver and egg) are given in **Table 41**. The mean As concentration in bovine milk collected from the exposed area is 6.37 ± 2.98µg/L (range: 3.29 to 13 µg/L), whereas that in the control site is 3.6 ± 1.12 µg/L (range: ≤ 3 to 5.08 µg/L). Datta et al. (2010) reported that cow milk consists of mainly i-As.

Table 41: Arsenic accumulation in different edible animal products from exposed zone

Animal Products	n	Mean	SD	Range
Cow milk (µg/L)	26	6.37	2.98	3.29-13.0
Milk collected from the cans of milk men (µg/L)	20	7.5	1.36	6.12-9.51
Goat milk(µg/L)	10	<3	-	-
Boiled Egg White (µg/kg)*	10	17.7	7.7	9.7-30.6
Boiled Egg yolk (µg/kg)*	10	18.6	7.62	9.78-22.1
Chicken meat (µg/kg)**	10	94.5	40.4	51.2-163
Chicken liver (µg/kg)**	8	192	80.4	118-328
Goat Meat (µg/kg)**	10	107	15	89-124

*wet weight, **dry weight

The methylated As from blood usually doesn't go into the epithelial cells of mammary glands. It is proved as all the goat milk samples in our study had <3 µg/LAs concentration. The boiled albumen and yolk has fairly comparable As concentration (mean: 17.7 ± 7.7 µg/kg and 18.6 ± 7.6 µg/kg). The range of weight of a boiled egg is between 24.6 to 54.2 g. So, the mean As content in a whole egg is 809 ng (range: 423-1519 ng). Liver sample of hen has higher Asconcentration (192 ± 80.4 µg/kg) than its flesh (94.5 ± 40.4 µg/kg). Similarly, goat flesh is found with substantial As concentration (range: 89-124µg/kg; mean: 107 ± 15 µg/kg). However, the acceptable concentration in egg and meat is 100 µg/kg (JECFA, 2005). **Table 42** gives idea about the health status of the heifers and accretion of As in consumable animal products reported from other studies in Bangladesh and West Bengal.

Table 42: Arsenic accumulation in edible animal products and livestock health exposure from other studies reported in West Bengal and Bangladesh in comparison with the present study

Location	**Target Animal**	**As Intake** (Mean; Range)	**As excrete** (Mean; Range)	**As in consumable animal products** (Mean; Range)	**Reference**
Nadia, West Bengal	Cattle (n= 60)	Water: 34-57 µg/L	Urine:61-98 µg/L Dung: 258-580 µg/kg Tail Hair: 1628-4065 µg/kg	Milk: 47-72 µg/L	Datta et al. (2012)
	Poultry birds (n= 40)	-	Feathers: 346-485 µg/kg	Egg albumin: 28-62 µg/kg Egg yolk: 71-111 µg/kg Liver: 20-72 µg/kg Meat: 29-30 µg/kg	
Nadia, West Bengal	Goats (n= 30)	Water: 143 ± 40 µg/L; n=23	Urine: 291 ± 162 µg/L Dung: 1283± 323 µg/kg Hair: 771 ± 41 µg/kg	Meat: 186 ± 89 µg/kg	Rana et al. (2012)
Nadia, West Bengal	Goats (n= 30)	Water: 132 ± 41 µg/L	Urine: 205 ± 63.3 µg/L Faeces: 700 ± 161 µg/kg Hair: 850 ± 21 µg/kg	-	Rana et al. (2014a)
Nadia, West Bengal	Chicken (n= 30)	Water:122 ± 40 µg/L	Feathers: 385 ± 125 µg/kg Litters: 535 ± 101 µg/kg	Whole egg: 222 ± 120 µg/kg Yolk: 107 ± 23 µg/kg Albumen: 65 ± 45 µg/kg	Rana et al. (2014b)
	Ducks (n= 30)	-	-	Whole egg: 155 ± 231 µg/kg Yolk: 46 ± 211 µg/kg Albumen: 95 ± 123 µg/kg	

North and South 24 Paraganas, West Bengal	Cattle	Water: 4- 488 µg/L (n= 89)	Urine: 51- 2424 µg/L; n= 93 Hair: 98- 2236 µg/kg; n= 138	-	Bera et al. (2010)
North 24 Paragans and Nadia, West Bengal	Cow	Water: 75 µg/L;3-746 µg/L; (n= 25)	Hair: 461- 984 µg/kg; n= 157 Urine: 2084 µg/L; 242-4643 µg/L; n=26 Dung: 6073 µg/kg; 3012-8997 µg/kg; n= 25	-	Pal et al. (2007)
Bangladesh	Cow (n=240)	Water: 89.6 ± 6.5 µg/L	Urine: 123.6 ± 7.6 µg/L Dung: 1693 ± 65.1 µg/kg	Milk: 26.2 ± 2.8 µg/L	Ghosh et al. (2013)
North 24 Paragans, West Bengal	Cattle (n=30)	Water: 86.7 ± 109 µg/L; 4.8-403 µg/L	Urine: 9.53 ± 7.37 µg/L; <3-28.2 µg/L Faeces: 794 ± 425 µg/kg; 237-1804 µg/kg Tail hair: 698 ± 356 µg/kg; 175-1491 µg/kg	Cow milk: 6.37 ± 2.98 µg/L; 3.29-13 µg/L	Present study
	Chicken (n=10)	Do	-	Egg: 36.3 ± 14.5 µg/kg; 22.5-59.6 µg/kg Liver: 192 ± 80.4 µg/kg; 118-328 µg/kg (n=8) Meat: 94.5 ± 40.4 µg/kg; 51.2-163 µg/kg	
	Goats (n=10)	Water: 309 ± 273 µg/L; 18-604 µg/L	Urine: 205 ± 108 µg/L; 92-338 µg/L	Milk: <3 µg/L (n=10); Meat: 107 ± 15 µg/kg; 89-124 µg/kg	

The milk samples collected from the cans of milk-men from the exposed site showed a slight higher As concentration (mean: 7.5 ± 1.36 µg/L). The most likely reason after this is the familiar practice of the milk-men of mingling water in raw milk to surge the volume. As-contaminated water upsurges the As concentration in milk. In the exposed cattle populace, milk As is majorly ruled by corn husk (r=0.845) and mustard cake (r= 0.842) (**Table 43**). Though, the input of As concentration in rice straw and drinking water on milk seems to be non-significant (r= -0.06 and -0.29). In the control population, rice straw and other grains may be accountable for As concentration in milk although the correlation values do not exhibit much significance (r= -0.45 and -0.018). Drinking water As has no such effect on milk, as explained by its reduced correlation factor (r = -0.29 and 0.012) from both exposed and control site. The standard As concentration in cow milk is 10 µg/kg (IDF, 1986; Sigrist et al., 2010). By FSSAI, the recommended As concentration in bovine milk is 100 µg/L (FSSAI, 2006; Sarkar et al., 2016), which displays that the cow milk As

concentration is under allowable limit. Children generally suffer from undernourishment in rural Bengal (Roychowdhury, 2010). Cow milk is provided to the babies starting from 12 months as one of the primary health drinks (Benelam et al., 2015). Thus, the smallest As contamination in milk also can produce health perils.

Table 43: Effect of arsenic in cow milk from various dietary sources based on Pearson's r correlation

Milk	Drinking water	Paddy straw	Corn husk	Mustard cake	Mixed commercial food
Exposed	-0.290 ***	-0.062**	0.845 **	0.842 **	NA
Control	0.012**	-0.457 *	NA	NA	-0.018 **

* p <0.05; **p <0.01; *** p≺ 0.05 or 0.01

5.5.8.1. Arsenic exposure in the fractions of whole milk

Cow milk consists of water, fat, lactose, whey proteins, casein, vitamins and minerals (Kennelly et al., 2000). Whole milk mainly has two parts, solid and water (by 12.6 and 87.4% respectively). The solid part contains lactose, fat, casein, ash and whey proteins by 38.1, 29.4, 22.2, 5.56 and 4.76% respectively (Chandan, 1997). This work involves estimation of As in the segments of whole cow milk in 16 chosen samples collected from the exposed area. The range of total As concentration in these milk samples are 9 to 13 µg/L with a mean value of 9.83 ± 1.6 µg/L. Arsenic concentration in different fractions of raw milk states that maximum accumulation took place in casein followed by fat, whey proteins and skimmed milk (83, 10, 4 and 3% respectively), which is shown in **Fig. 52**. Chandan (1997) stated that casein carries almost 80% milk proteins and the remaining is insoluble whey proteins. Proteins largely make milk casein (Cn); αs1-Cn, αs2-Cn, β-Cn and k-Cn coupled to several phosphate groups and no disulfide bonds (Andrei 2006). Bhat et al. (2016) and Walstra et al. (1999) identified that the proteins αs1-Cn and αs2-Cn contain 8 to 13 phosphoserine groups, while, β-Cn and k-Cn proteins have respectively 5 and 1 to 2 seryl phosphate moieties. Phosphoserine is an ester formed by serine and phosphoric acid, known as predecessor of peptides that contain binding sites for minerals, as reported by Peres et al. (1997) and Vegarud et al. (2000). Incidence of highest% of As in casein is a probable result of metal entrapping in these groups as it is apparent that phosphoric acid acts like a chelating agent. Bundschuh et al. (2012) and Vahter and

Concha (2001) reported As accumulation in casein of cheese (0.42 ± 0.04 mg/kg) during milk curdling (0.44 ± 0.06 mg/kg). Daus et al. (2006) also showed that phosphoric acid is used to conserve As species in water for long.

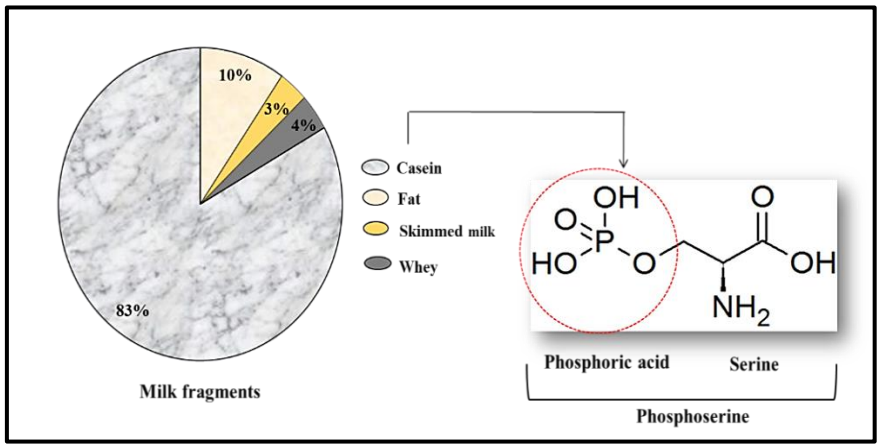


Fig. 52. Arsenic accumulation in different fractions of whole cow milk

5.5.8.2. Arsenic accretion in the by -products of milk

Milk based items had been collected from the local sweet shops in the exposed sites to quantify the added As burden caused by As-contaminated milk (**Fig. 53**). A substantial amount of mean As is noticed in each components of sweets like curdled milk (72.2 µg/kg), curdled milk-water (11.1 µg/L), sweets (79.8 µg/kg) and sweet syrups (52.2 µg/L) made from raw milk (8.06 µg/L). The sweets collected in this study are natively called 'rasogolla' which is usually dipped in a sugar-syrup for a long period prior to selling. Possible reason for elevated As in sweets is the concentration of As in the water used for preparation of syrup. Furthermore, the increased As concentration in milk by-products might be caused by reduction of volume during boiling of milk.

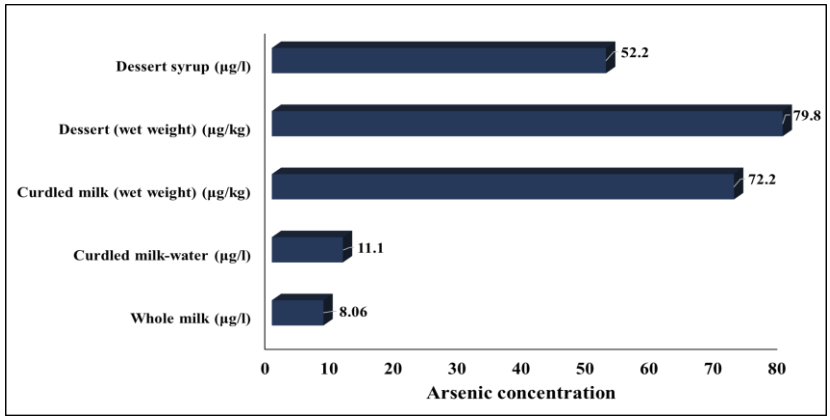


Fig. 53. Arsenic accumulation in milk by products from exposed areas (local shop-based study)

5.5.9. Association between arsenic intake and excrete

At 95% confidence level, a 'two tailed paired t test' has been performed to comprehend the reliance of excreted As from animal body with As intake through different intake routes. 'No significant dependence of excreted As with As intake' is considered as Null hypothesis (Ho) and, 'significant interdependence of both the variables' is taken as alternative hypothesis or H_1. **Table 44** elaborately showed the t values in each relation with relevant degrees of freedom (df). Significant relation is found among As concentration in drinking water and that in urine or milk in the exposed cattle population, whereas, excreted As is fully dependent on As concentration in paddy straw. As a result, Ho is discarded in the above cases. In the control cattle population, there is no significant link found among As concentration in drinking water and excreta samples, while significant dependence is found among As concentration in rice straw with that in urine and milk, indicating rejection of Ho. This is an obvious outcome of As free water in the control sites. Most often, the domestic livetsock in the unexposed area are treated with market feed which are transported from As endemic sites, reported in Biswas et al. (2019). Thus, rice straw is an obvious source of As toxicity in livestock. Surprisingly, Ho is accepted for drinking water and urinary As in case of the exposed goats, and rejected in paddy straw.

Table 44: 'Two tailed paired t test' analysis for health exposure of arsenic in livestock

Study Area	Arsenic concentration	Urine	Faeces	Tail Hair	Milk	Remarks (Null hypothesis)
Exposed (cattle)	Drinking water	t_{stat} (3.36) $>t_{critical}$(2.09) df = 25	t_{stat}(-9) $<t_{critical}$(2.09) df = 26	t_{stat} (-7.33)$<t_{critical}$(2.09) df = 26	t_{stat}(2.26) $>t_{critical}$(2.13) df =20	Rejected for urine and cow milk
	Paddy straw	t_{stat}(5.12) $>t_{critical}$(2.12) df = 25	t_{stat}(3.26) $>t_{critical}$(2.12) df = 26	t_{stat}(3.32) $>t_{critical}$(2.12) df = 26	t_{stat}(4.99) $>t_{critical}$ (2.15) df = 20	Rejected in each case
Exposed (goats)	Drinking water	t_{stat} (0.79) $<t_{critical}$(2.78) df = 12	-	-	-	Accepted
	Paddy straw	t_{stat}(3.25) $>t_{critical}$(2.78) df = 12	-	-	-	Rejected
Control (cattle)	Drinking water	t_{stat} (-2.83) $<t_{critical}$(2.36) df = 8	t_{stat} (-12) $<t_{critical}$(2.36) df = 8	t_{stat}(-3.4) $<t_{critical}$(2.36) df = 8	t_{stat}(-1.42) $<t_{critical}$(2.45) df = 7	Accepted in each case
	Paddy straw	t_{stat}(4.19)$>t_{critical}$(2.36) df = 8	t_{stat}(1.49)$<t_{critical}$(2.36) df = 8	t_{stat}(0.83)$<t_{critical}$(2.36) df = 8	t_{stat} (3.55)$>t_{critical}$(2.45) df = 7	Rejected for urine and cow milk

5.5.10. Exposure of arsenic and application of Risk thermometer

Arsenic exposure per day (µg/kgbw/day) is assessed in livestock and humans to realize the probable health adversity. As a result, the health risk in humans from the food products has been anticipated by an appliance of 'Risk Thermometer' (eq. 14) (**Fig. 54**).

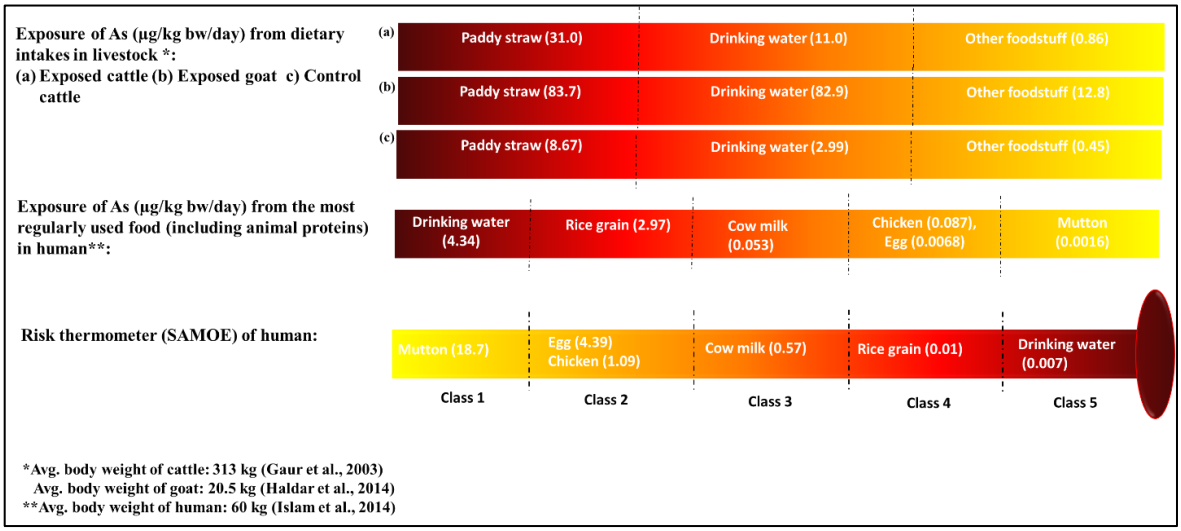


Fig. 54. Exposure of As in livestock and human through edible animal products in human population based on SAMOE calculation

It is found that the domestic livestock from both the study areas are affected with As through rice straw then by drinking water and other products. Although, the exposure is lesser in the control populace than the exposed cattle populace. When the animal sourced proteins are considered, the maximum chance of As mediated toxicity in humans comes from bovine milk after that chicken, egg and goat flesh. In addition, when drinking water and rice grain are considered, health risk rises considerably. A consumption of 425 g rice grain per day (As concentration: 419 µg/kg) and 3 L drinking water (As concentration: 86.7 µg/L) gives higher health risk (Class 4 and class 5) I comparison to the animal proteins (**Table 45**). Fig. 54 explains that cow milk gives low to moderate risk (class 3), while both egg and chicken flesh give no to low order of risk (class 2). Consuming goat flesh apparently produces no health risk as it comes in class 1. Consumption of minimum quantity of As via cow milk for an extended time can therefore fetch a future life risk.

Table 45: Calculated SAMOE values and risk level for an arsenic exposed adult

Animal products	Mean As concentration	Daily ingestion rate (a, b, c)*	Daily intake (µg/day)	Daily Exposure (µg/kg bw/day) (BW= 60 kg)**	SAMOE	Risk level
Goat meat or mutton (µg/kg)[a]	107	0.9 g	0.0963	0.001605	18.69159	Class 1
Chicken meat (µg/kg)[a]	94.5	17.4 g	1.6443	0.027405	1.094691	Class 2
Egg[a] (µg/kg)	36.3	11.3 g	0.41019	0.006837	4.38821	Class 2
Milk[b] (µg/L)	6.37	0.5 l	3.185	0.053083	0.565149	Class 3
Rice Grain (µg/kg)[c]	419	425 g	178	2.97	0.010101	Class 4
Drinking water (µg/L)[c]	86.7	3 l	260	4.34	0.006912	Class 5

a** BBS (2011); Shaheen et al. (2015),b** Kazi et al. (2016), **c** Rahman et al. (2013); **Islam et al. (2014)

5.5.11. Cancer and non-cancer risk assessment

Lifetime cancer and non-cancer risk are estimated for humans based on an estimated average daily dose of the contaminant, which is called ADD (USEPA, 1998) (eq. 10-12).

C is As concentration in the food objects; IR is the Rate of ingestion; ED is duration of exposure (ED_{Adult} = 70yrs, ED_{Child}= 5 yrs); EF is Exposure frequency (365 days/yr); BW is Body weight (BW_{Adult}=60 kg, BW_{Child}= 16 kg) (Islam et al., 2014, 2017) and AT is Average Lifetime (25,550 days).

It tells that the adults have tendency to As toxicity than children (**Table 46**). The cumulative cancer risk value ($\sum CR_{adult}= 1\times10^{-2}$, $\sum CR_{child}= 9\times10^{-4}$) is surprisingly higher than the threshold value, where contribution of chicken, egg and cow milk is not insignificant. The HIchild and HIadult are 2.01 and 22.9 that cross the threshold limit. Rice grain-HQ is significant only in adults with a value of 9.19 whereas that of drinking water is significant in both child and adults (1.94 and 13.4 respectively).

Table 46: Human health risk from most consumed daily diets in arsenic exposed areas

Animal products	Mean As concentration	Daily ingestion rate (a, b, c)*		Average Daily Dose (ADD) (mg/kg/day)		Non-cancer Risk (HI)		Cancer Risk (CR)	
		Child **	Adult	Child	Adult	HQ_{Child}	HQ_{Adult}	Child	Adult
Goat meat or mutton (µg/kg)[a]	107	0.3 g	0.9 g	1.43×10^{-7}	1.60×10^{-6}	0.0005	0.005	2.15×10^{-7}	2.40×10^{-6}
Chicken meat (µg/kg)[a]	94.5	8.3 g	17.4 g	3.5×10^{-6}	2.74×10^{-5}	0.012	0.09	5.25×10^{-6}	4.11×10^{-5}
Egg[a] (µg/kg)	36.3	6.5 g	11.3 g	1.05×10^{-6}	6.83×10^{-6}	0.004	0.022	1.58×10^{-6}	1.02×10^{-5}
Milk[b] (µg/L)	6.37	0.25 l	0.5 l	7.11×10^{-6}	5.30×10^{-5}	0.023	0.18	1.07×10^{-5}	7.39×10^{-5}
Rice Grain (µg/kg)[c]	419	280 g	425 g	5.24×10^{-4}	2.96×10^{-3}	0.039	9.89	1.77×10^{-5}	4.45×10^{-3}
Drinking water (µg/L)[c]	86.7	1.5 l	3 l	5.81×10^{-4}	4.33×10^{-3}	1.94	14.5	8.7×10^{-4}	6.5×10^{-3}
Total	-	-	-	-	-	**HI= 2.01**	**HI= 24.6**	**$\sum$CR =** 9×10^{-4}	**$\sum$CR =** 1×10^{-2}

a**BBS, (2011); Shaheen et al., (2015);b**Kazi et al., (2016); **c** Joardar et al., (2021a), Rahman et al.,2013**(Child age: ≤ 5 years)

5.5.12. Environmental risk: Bio-transformation factor (BTF)

The environmental risk is calculated through biotransformation factor (BTF). It is an approximation to perceive how much contamination is biologically transferred in the environment from the animal bodies (Rana et al., 2014a; Liao et al., 2008).

$$\text{Biotransformation factor} = \frac{\text{As concentration in excretion samples}}{\text{As consumption only through drinking water}} \quad \text{eq. (18)}$$

Arsenic content of livestock excreta brings a considerable environmental risk, being accumulated in soil readily, that results into the enrichment of in-situ soil As. Thus, As is translocated in plant bodies, taken by the animals and the chain continues. Cultivated grains are transported to adjacent and farther urban areas, and the As exposure extents a health risk to the apparently unexposed populace (Biswas et al., 2019). Cattle dung, a conventional bio-resource is used as a fundamental constituent of agrarian farm yard management, fuel component in village or mosquito repugnant (Gupta et al., 2016; Rana

et al., 2014a). Pal et al. (2007) stated that burning of fuel with contaminated faeces can be a prospective mediator of indoor air pollution. The computed bio-transformation factor follows the order: BTF_{dung}> $BTF_{tail\ hair}$> BTF_{urine}> BTF_{milk} in exposed cattle, and $BTF_{tail\ hair}$> BTF_{dung}> BTF_{urine}> BTF_{milk} in the control populace (Fig.51b). The pattern indicates that excretion of As from cattle's body occurs through faeces at a higher rate than tail hair and urine (Fangstorm et al., 2008; Ghosh et al., 2013). The biotransformation rate is greater in control cattle populace than the exposed that might be due to their virtuous metabolic structure.

A case study was performed to check whether the cow dung As concentration enhances the soil As or not. Arsenic concentrations in cow dung, cow shed soil and fallow land soil were analysed for both exposed and control subjects (**Fig. 55**). It showed that in the As exposed area, As concentration was highest in cowshed soil (average: 2461 µg/kg, range: 2032-2917 µg/kg) followed by cow dung (average: 871 µg/kg; range: 413-1789 µg/kg) and fallow land soil (average: 508 µg/kg; range: 326-784 µg/kg) (Fig. 55a). This proves that the As from animal excreta (faeces) gets mixed into the nearest soil and increases the soil's indigenous arsenic concentration. So, the plants grown in the nearby soil is expected to experience As stress at a relatively higher rate. At the same time, As concentration was highest in cowshed soil (average: 377 µg/kg; range: 228-538 µg/kg) followed by fallow land soil (average: 329 µg/kg; range: 205-422 µg/kg) and cow dung (average: 160 µg/kg; range: 113 - 222 µg/kg) (Fig. 55b). Accumulation of As in water, fodder and excreta is less in control population than exposed which leads to lower accumulation in cowshed soil.

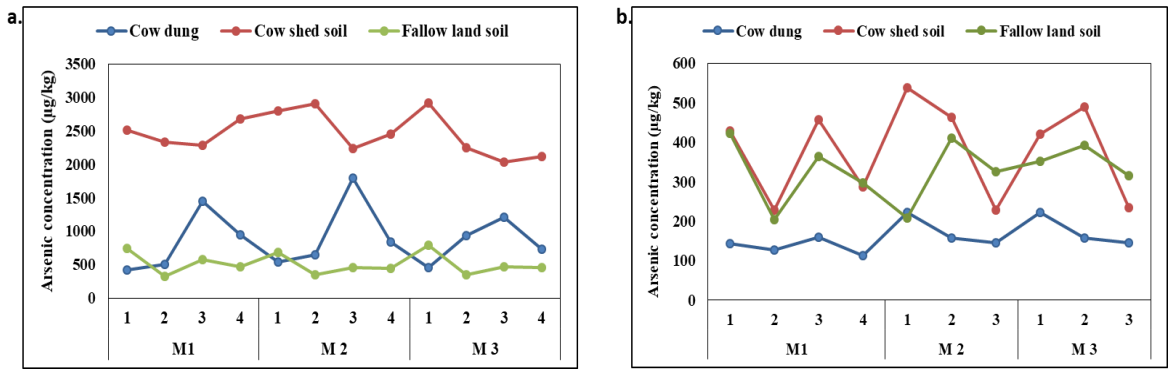


Fig. 55. Month-wise arsenic accumulation in cow dung and nearby soil: (a) exposed area, (b) control area

The livestock suffers from sub-clinical As toxicity and causes a health risk in humans through the animal sourced goods. Arsenic concentrated animal excrements get added with soil ensuing low soil fecundity and inferior crop quality. This cycle stimulates bioaccumulation and bio-magnifications as the metal goes into plant, followed by animal bodies. Arsenic gets transferred from one trophic level to another through the food chain and humans get affected via maximum routes (drinking water, raw and cooked food especially rice, animal proteins etc.). So there is a definite possibility of bio magnification of As in human tissues which is a major topic of research in future. In terrestrial ecosystems, bio-magnification of trace elements is known along food cycles, as consecutive trophic levels devour more food or biomass to fulfill their metabolic functions (Rehman et al., 2021).

5.6. Mitigation options: Alternate drinking water sources

West Bengal government and Bangladesh launched a two phase program to alleviate Ascontamination in groundwater of several As affected areas in association with the World Health Organization (WHO), United Nations Children's Fund (UNICEF), World Bank, and various international agencies and Non-Governmental Organizations (NGOs) as a part of their mitigation strategy (Hossain et al., 2006a,b). They identified the As contaminated shallow tube-wells (>50 μg/L) and marked them 'red' while they distinguished the lower Asconcentrated tube-wells 'green'. Several efforts have been made from the government and non-governmental organizations to provide safe drinking water. Short term measures for community level plants involve technology models developed by several National and International agencies. For household level, available technologies are like Filter Tablet system, developed by School of Environmental Studies (SOES), Jadavpur University, AIIH&PH and the third and last one is National Metallurgical Laboratory (NML), Jamshedpur. It is recommended that surface water, flooded river basins, village ponds, dug wells and rainwater are existing in abundance in our state and may be harnessed for having As free drinking water. Arsenic removal from groundwater depends on several factors like pH of the medium, valence state of As, redox potential, presence of other cations and anions etc. Oxidation, precipitation, adsorption, coagulation and flocculation, ion exchange, membrane filtration etc. are the various methodologies available for As removal from groundwater. Phytoremediation, microbial remediation, chemical precipitation, electro-kinetic methods are also conventionally known methods for the same. At recent past, many of the technologies are effectively formed and performed in Bengal delta. Few technologiest hrived well in the laboratory but they are not fruitful in practical life. A number of community level As removal technologies were established in West Bengal by several national and international agencies based on different scientific principles, e.g. 'Apyron technology' using enhanced manganese oxide and activated alumina adsorption media, 'Water Systems International technology or WSI' using bucket of resin or ion exchange method, 'RPM technology' using activated alumina, 'Pal Trockner – AdsorpAs technology', 'Anir Engineering' using Granular Ferric Hydroxide (GFH), 'School of Fundamental Research (SFR) technology' containing PVC cylinders packed withgoethite specifically deposited on activated alumina, 'PHED technology' using sand-activated

alumina, ‘AIIH & PH’ or All India Hygiene and Public Health technology and Oxide India (Catalysts) Pvt. Ltd. (B.E. college model) etc. (Bhavan, 2007). Reports showed that many of the plants’ efficiency decreases within few years of establishment in India as well as Bangladesh and Nepal (Kabir and Howard, 2007; Gebauer and Saul, 2014). On January, 2006, outcomes of the examination of 12 Arsenic removal plants set at the technology park, Baruipur in West Bengal through the “Report of the task force on formulating action plan for removal of As contamination in West Bengal” stated the most of the technologies are either not functioning or missing or not existing. Even Das et al. (2016) reported that almost 95% As mitigation technology in several affected districts of this state became non-functional within one year of installation. In many instances, families are not using the treatments due to several practical reasons especially lack of willingness to pay (Dasgupta et al., 2022); due to negligence and incognizance as they undervalue the welfares of As remediation as it is invisible and unscented in drinking water and its adverse effect is not instant (Gebauer and Saul, 2014; Roy, 2008). Therefore, an “Arsenic Master Plan” wasset by the Public Health Engineering Department or PHED, Govt. of West Bengal with collaboration to Arsenic Task Force in 2018 suggesting ways for the provision of safe drinking water to the As affected community through building tube-wells and Piped Water Supply Schemes (PWSS).

The available alternate drinking water sources include arsenic iron treatment units (ATU), Sajaldhara treatment plant, reverse osmosis plants and supply of treated surface water (pond or river) by pipeline, dug wells etc. There is no denial of the fact that the drinking water through these alternate sources contains adequate As concentration to cause substantial health risk. The scenario in the studied sites of Murshidabad is illustrated in the following table (**Table 47**).

Table 47: Arsenic concentration in alternate drinking water sources of Murshidabad

Water source	Depth (ft)	Arsenic concentration			Iron concentration		
		Number of sample (n)	Mean (µg/L)	Range (µg/L)	Number of sample (n)	Mean (mg/L)	Range (mg/L)
Domkol block, Murshidabad							
PHED supplied tap water in the municipal area (tank source)	-	2	13.4	11 – 16.9	1	0.85	0.1 – 1.6
PHED deep pump	70	1	36.2	-	1	0.7	-
Raw water through deep pump	220-240	4	80	30-158	2	0.95	0.5 – 1.4
Filter water (50 jar sold/day; 20 l/jar)	-	4	10.7	<3 – 22.6	-	-	-
Tube well drinking water (sold 20-25 jar/day; 20 l/jar)	240	2	<3	-	2	0.05	-
Raninagar II block, Murshidabad							
Pipeline supplied water	120 – 170	10	21	<3-48.1	10	1.27	<0.05- 4.7
Dug well	30 – 70	5	3.78	<3-5.5	5	0.17	<0.05- 0.36
Sajaldhara-Arsenic Removal Plant (Raw)	-	6	69.6	34.7-136	6	2.05	1.74- 6.5
Sajaldhara-Arsenic Removal Plant (Treated)	-	16	13.6	<3-56.1	15	1.40	<0.05- 4.31
Reverse Osmosis (R.O.) Arsenic Removal Plant (Raw)	-	4	98.8	12.9-274	4	2.70	0.92- 6.37
Reverse Osmosis (R.O.) Arsenic Removal Plant (Treated)	-	5	7.76	<3-17.4	5	0.06	<0.05-0.1
Chakdah block, Nadia							
Treated surface water through Pipeline	-	8	3.49	<3 – 8.4	7	2.02	0.1 – 9.46
Arsenic removal plant treated water (filter)	-	3	<3	-	3	0.1	-

5.6.1. Pipeline

Till date, 23,054 villages in West Bengal are covered with commissioned PWSS which includes a total rural population of 4,64,56789 and 147 schemes are on the process (www.wbphed.gov.in). The pipeline supplied water is mainlypumped out from the river Bhagirathi or Ganges and treated in surface water treatment plantsby standard treatment processes; alum dosing for coagulation and turbidity removalfollowed by flocculation and clarification, sandbed filtration and lastly, chlorination for disinfection (Biswas et al., 2021). The local populations depend on the supplied pipeline water through Public Health Engineering Department (PHED), which is not available throughout the study area. Consequently, the populationhas to rely on domestic and community level shallow tube-wells. It seems that there exists a gap between water supply system and community welfare of availability of pure water. In the studied sites of Murshidabad, it is seen that the source of available pipeline water is nothing but thegroundwater in the depth range of 120-170 ft. which is supplied after direct withdrawal using motor pump and sometimes stored inreservoirs for overnight prior to supply twice a day for few hours. In Nadia (Chakdah block), the pipeline supplied water is Ganges treated water; average As concentration is 4.15 μg/L, (n = 7). The pipeline supplied water in studied Raninagar II and Domkol blocks shows average As concentration of 21 and 13.4 μg/L (range: <3–48.1 μg/L; and11 – 16.9μg/L) which is unsafe for drinking. Nearly, 83 % of the analyzed samples (n = 12) in Murshidabad have been found with As concentration above permissible limit. The scenario is verified through similar kind of findings by Mazumder et al. (2020) in Maldah district, West Bengal. They observed that out of the studied 81 habitations, 68 had pipelines built by PHED for supply of As free drinking water. Nevertheless, only 4 habitations had regular water supply, 38 had irregular supply and 26 had no supply at all. Due to inadequacy of As free pipeline supplied water, people were forced to drink water from PHED tube-wells where it was seen that 76% samples were contaminated with As greater than the recommended limit of drinking water. In addition to this, their report on illegally tapping of pipeline is frequent in these rural beltsto avail water for various purposes which eventually cause break of pipelines during water supply by PHED. Under the Jal Jeevan Mission (JJM), States and UTs have been suggested to plan and implement mass water-transfer schemes using piped water supply schemes that are based on safe water sources, such as surface water sources or alternative safe ground water sources for the villages with poor water quality. States or UTs have been recommended to establish community water

purification plants (CWPPs) primarily in As -affected populations to offer drinkable water to the households at 8-10 l per capita per day (lpcd) to meet their daily requirements (drinking and cooking) because planning, implementing, and authorizing a safe water piped water supply scheme may take time. Remedial actions including PWSS of treated surface water and harvested rainwater are definitive ways to achieve sustainable de-arsenification; although these ways can furnish to a limited population having logistic and monetary restraints. The establishment of ATUs, that treat extracted underground water to provide safe drinking water, would persist as the foremost remedy to remediate As in the far-flung rural habitations of our country.

5.6.2. Arsenic removal plants

In few studied blocks of Murshidabad, it is found that in few places, the pipeline supplied water and the treated water produced from the arsenic removal plants contain adequate As to cause health risk. In the present research study, in Raninagar II, Murshidabad, it is observed that in the treated water from Reverse Osmosis (R.O.) plants, about 20% of analyzed samples were above permissible limit of As in drinking water while in case of Sajaldhara, it was 31.3%. Arsenic concentration in the raw water samples of R.O. plants and Sajaldhara plants was (range: 12.9 -274 µg/L, average: 98.8 ± 103 µg/L, n= 4) and (range: 34.7-136 µg/L, average: 69.6 ± 36 µg/L, n= 6). Whereas, As concentration in the treated water samples of R.O. plants and Sajaldhara plants was (range: <3-17.4 µg/L, average: 7.76 ± 5.26 µg/L, n= 5) and (range: <3-56.1 µg/L, average: 13.6 ± 16.8 µg/L, n= 16). Arsenic removal efficiency of the R.O. plants and Sajaldharawater treatment plants were 77.6% and 74.4% respectively. Fe concentration range inraw and treated water of R.O. plants are 0.92–6.37 and < 0.05–0.1 mg/L, respectively. However, the mean Fe concentration value in treated water by Sajaldhara plants (1.27 mg/L; range: <0.05–4.31 mg/L) is still 4.23 times higher than its recommended value in drinking water.

5.6.2.1. Performance evaluation of few selected arsenic treatment plants in severely arsenic exposed villages of North 24 Parganas, West Bengal

In many areas, people still use the As contaminated tube-wells due to lack of awareness or insufficient access of the safe water. In addition to that, poor efficiency of the ATUs installed in West Bengal compels the people to imbibe As-contaminated water. Murphy et al. (2010) reported that the As treatment units established in rural Bangladesh got a high social acceptance, whereas with time it has been seen that over 70% plants had been

abandoned as well as the As removal percentage got decreased. Sorenson and Mcbean (2015) discussed about the social considerations for the sustainable use of ARPs in Bangladesh where they reported the number of possible reasons behind the discontinuation in usage of the plants. The total number of sanctioned arsenic-iron removal plants in West Bengal is 187 among which 139 are already commissioned and 49 are in process (www.wbphed.gov.in). The number of commissioned As removal plants in Maldah, Murshidabad, Nadia and North 24 Parganas districts are 11, 31, 71 and 26 respectively. Conventionally, the ARPs avail several methods for de-arsenification of contaminated water including co-precipitation, oxidation, coagulation, lime treatment, adsorption, ion-exchange resin and membrane techniques etc. (Ahmed et al., 2001). But, ion exchange or membrane technology are comparatively expensive methods and not pertinent in the scenario of West Bengal. A few of these technologies can be condensed in size and be applieddomestically for removal of Asfrom contaminated water. The most commonly used process of As removal from contaminated groundwater is co-precipitation through use of naturally occurring iron and adsorption in different media. The iron precipitates [Fe $(OH)_3$] produced by oxidation of dissolved iron in groundwater have the affinity for As adsorption. Generally, the ARP is a unit which consists of two tanks that work on oxidation and co-precipitation. The larger oxidation tank and the small co-precipitation tank have coarse sand, charcoal, and brick chip media (Rahman et al., 2021; Sorenson and Mcbean 2015). Koley (2020) and Koley (2021) has been discussed the pros and cons of the groundwater As remediation strategies in West Bengal through As removal plants and units with detailed description of the technologies involved. The article has also described the scenario of wastewater and sludge management processes.

5.6.2.2. Description of the selected ARPs in the study area

Groundwater of North 24 Parganas district, West Bengal is severely As contaminated (Roychowdhury, 2010; Joardar et al., 2021a) and natural groundwater contains a huge amount of iron. The scattered As removal plants are the most plausible option to provide arsenic-safe drinking water. The study initially carried out with estimation of As concentration in the treated water of 24 ARPs which revealed that not all the units are well maintained. The details of the plants including their geographical location and source of raw water are given in **Table 48**.

Table 48: Details of the arsenic removal plants

ARP no.	GPS		Location	Source of raw water	Arsenic concentration in treated water (µg/L)
	Latitude	Longitude	Name of G.P		
1#	22°53′5106″	88°46′3807″	Sutia	Groundwater	10
2	22°54′0336″	88°47′1150″	Sutia	Groundwater	7.35
3#	22°53′5120″	88°46′3812″	Sutia	Surface water	7.74
4#	22°55′55.69″	88°45′2680″	Ichapur-I	Groundwater	<3
5#	22°55′22.64″	88°45′37.89″	Ichapur-I	Groundwater	70
6#	22°53′50.19″	88°46′0925″	Ichapur-II	Groundwater	191
7	22°54′0160″	88°44′1045″	Ichapur-II	Groundwater	126
8#	22°57′25.06″	88°47′03.58′′	Chandpara	Groundwater	21.6
9#	23°00′0355″	88°46′4684″	Chandpara	Groundwater	38.6
10#	22°59′0473″	88°48′1593″	Dooma	Groundwater	18.5
11	22°57′16.74″	88°48′52.16′′	Dooma	Groundwater	117
12	22°56′3888″	88°50′0312″	Jhaudanga	Groundwater	20.5
13	22°56′5622″	88°50′0151″	Jhaudanga	Groundwater	21.7
14#	22°55′34.41″	88°48′10.50″	Shimulpur	Groundwater	29.2
15#	22°55′5166″	88°49′5894″	Shimulpur	Groundwater	120
16	22°56′0162″	88°43′2746″	Jaleswar-I	Groundwater	432
17#	22°56′02.21″	88°43′04.59″	Jaleswar-I	Groundwater	108
18	22°57′1134″	88°43′5805″	Jaleswar-II	Groundwater	59.4
19	22°57′1030″	88°43′6080″	Jaleswar-II	Groundwater	118
20	22°53′5936″	88°42′2207″	Dharampur-I	Groundwater	7.59
21#	22°54′5930″	88°42′0405″	Dharampur-I	Groundwater	65.2
22	22°55′0952″	88°41′5811″	Dharampur-II	Groundwater	11.9
23	22°52′2771″	88°40′1071″	Dharampur-II	Groundwater	46.7
24	22°55′4824″	88°52′3007″	Ramnagar	Groundwater	8.85
25	22°55′5763″	88°51′1091″	Ramnagar	Groundwater	6.98

 # Selected ATUs for the detailed study

48 The typical mechanism of the ATUs run in Bengal delta are already described in Hossain et al. (2006a) and Basak et al. (2021). There may be variation in the chemicals used for adsorption media, water softening or disinfection. Also, few plants contain iron guard, few plants don't. In the present work, among the 25 studied ATUs, 12 ATUs were selected for a one-year study to evaluate their efficiency. Images of 4 plants are shown below (**Fig. 56).**

Fig. 56. Studied selected arsenic treatment units in North 24 Parganas

The study reports that all the ARPs used raw groundwater and performed some basic treatment methods to attain safe drinking water. Plant no. 3 is a surface water treatment plant and the detailed technology for the mentioned water treatment plant is shown in **Fig. 57.** Sulabh International Social Service Organization (SISSO), in partnership with a French company, 1001 Fontaines, established a Rs. 20-lakh pond-based water treatment plant in Madhusudankati village of North 24 Parganas district, which is maintained by Madhusudankati Krishak Kalyan Samity' (a local non-Government organization). The treated water of this plant is bottled in a 20 L jar ('Sulabh Jal'), sold to the neighboring villagers and provided at free of cost to the chronic As patients (**Fig. 58**).

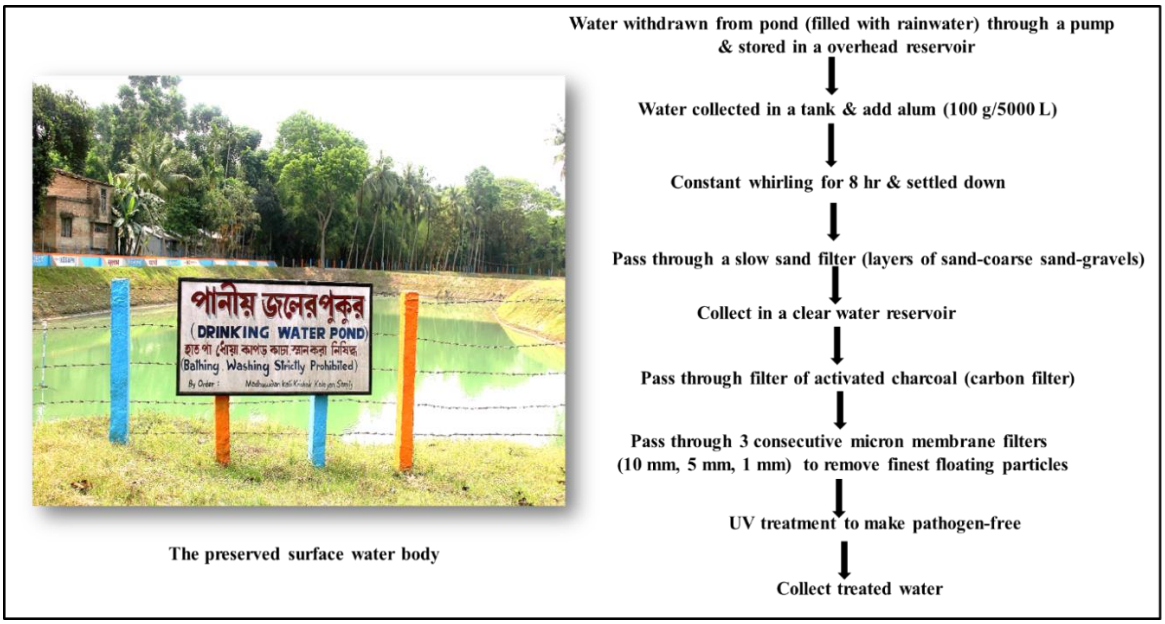


Fig. 57. Flow diagram for the mechanism of the surface water treatment plant (ARP no. 3)

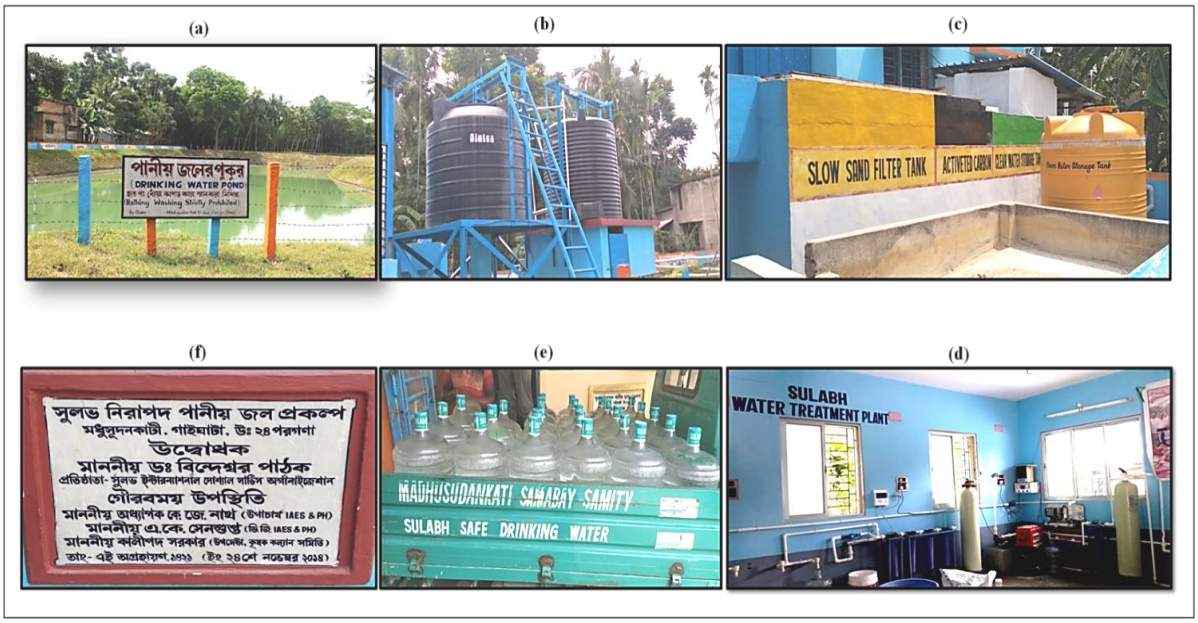


Fig. 58. Flow diagram of Sulabh water treatment plant procedure

5.6.2.3. Physicochemical parameters of raw water of the selected ARPs

The water quality is analyzed through estimation of 15 physicochemical parameters of raw groundwater samples (n= 12). **Table 49** gives the idea on the statistical arrangement of the water quality parameters of the samples. The color of the raw groundwater samples is observed to be clear except that of plant no. 3 and 15 which are slight brownish. The range of pH, EC and turbidity of the groundwater samples is 7.37 -8.0 (average: 7.60), 176 – 8330 µS/cm (average: 3666 µS/cm) and 9 – 35 NTU (average: 17.9 NTU). This suggests that the quality of the raw water of the plants is neither acidic nor alkaline in nature, not turbid but has a very much good potential to carry minerals. Singh et al. (2016) reported the hydro-geochemistry of North & South 24 Parganas districts with an average pH of 7.27 (range: 6.8-8.1), EC of 869 µS/cm (range: 540-1300 µS/cm) in groundwater samples. The average TDS and TSS value of the raw water samples are 606 and 698 mg/L which are slightly higher than the recommended value in drinking water, so it needs to be lessened through proper treatment. TDS value is exceptionally high in the raw groundwater sample of the treatment plant no. 1 (1733 mg/L). Higher TDS in groundwater is usually not detrimental to human life but may distress thepersons who suffer from kidney and heart maladies (Geetha et al., 2008). The range of total alkalinity (150 – 600 mg/L; average: 448 mg/L) and total hardness (60 – 390 mg/L; average: 300 mg/L) in the samples recommends that hardness and alkalinity need to be reduced prior to reach the population. The average concentration of the cations in the raw water samples are 51 mg/L (range: 20-80 mg/L),

55.09 mg/L (range: 24.3 – 70.6 mg/L), 33.5 mg/L (range: 11 – 123 mg/L) and 6.47 mg/L (range: 2.9 – 15 mg/L) respectively for calcium, magnesium, sodium and potassium (Table 2). Average concentration for the anions (chloride and sulphate) are 44.8 mg/L (range: 17.8 – 117 mg/L) and 3.98 mg/L (range: 0.9 – 15 mg/L) respectively (Table 2). The cumulative mean order of the ionic concentrations is in the order of Mg^{2+}> Ca^{2+} > Cl^{-}> Na^{+}> K^{+}> SO_4^{2-}. Singh et al. (2016) showed similar type of groundwater ionic concentrations in the two 24 Parganas districts of Bengal; sodium (average: 85.27 mg/L, range: 48.5-173.7 mg/L), potassium (average: 3.85 mg/L, range: 1-8.20 mg/L), magnesium (average: 65.39 mg/L, range: 56.79- 88.61 mg/L) and calcium (average: 97.83 mg/L, range: 67.53-139.34 mg/L) respectively.

Table 49: Physicochemical parameters of raw water of the selected ARPs

	Parameters														
ARP no.	**Color**	**Odor**	**pH**	**EC (µS/cm)**	**Turbidity (NTU)**	**TDS (mg/L)**	**TSS (mg/L)**	**Total Alkalinity (mg/L)**	**Total Hardness (mg/L)**	**Cl^- (mg/L)**	**SO_4^{2-} (mg/L)**	**Ca^{2+} (mg/L)**	**Mg^{2+} (mg/L)**	**Na^+ (mg/L)**	**K^+ (mg/L)**
1	-	No smell	7.44	2560	12	1733	850	440	380	17.8	1.5	56	58.4	16.2	3.2
3	Slight brownish	No smell	7.86	8330	14	466	1350	530	380	117	2	36	70.6	123	4.1
4	-	No smell	7.87	5800	35	616	100	410	340	42.9	1.2	36	60.8	30.5	19
5	-	No smell	7.37	5060	9	533	1450	480	260	21.3	1.2	56	29.2	19.6	3.5
8	-	No smell	7.38	5090	9	500	750	460	370	24.9	15	52	58.4	21.9	3.6
9	-	No smell	7.58	2400	30	400	700	480	390	46.2	0.9	56	60.8	30.1	3
10	-	No smell	7.39	4680	17	566	150	540	310	21.3	10	28	58.4	19.5	3.6
15	Slight brownish	No smell	7.42	5930	15	400	450	460	330	42.6	5	20	68.1	29.9	3
17	-	No smell	7.5	769	12	666	1100	440	320	17.8	1	72	34.1	16.7	4.7
21	-	No smell	7.86	176	16	567	950	380	250	17.8	1	60	24.3	11	2.9
6		No smell	8	3000	26	550	320	600	210	68	3.5	80	68	35	15
14		No smell	7.5	200	20	280	200	150	60	100	5.5	60	70	48	12
Average	-	-	7.60 ± 0.23	3666 ± 2563	17.9 ± 8.34	606 ± 370	698 ± 462	448 ± 111	300 ± 94.7	44.8 ± 33.7	3.98 ± 4.41	51.00 ± 17.7	55.09 ± 16.4	33.5 ± 29.9	6.47 ± 5.57
Acceptable limit in drinking water			6.5-8.5	-	5	500	-	200	200	250	200	75	30	200	12

5.6.2.4. Physicochemical parameters of treated water of the selected ARPs

The percentage of increase or decrease in the analyzed physico-chemical parameters in treated water from raw water has been shown in **Fig. 59.** The treated water of the selected 12 ARPs show that pH has been reportedly decreased by 26.4 %, 23.4 %, 11.6 % and 13.3 % in the plants no. 4, 9, 15 and 14 respectively whereas, it has been increased by approximately 6 % in the plants no. 8 and 10 (Fig. 59a).

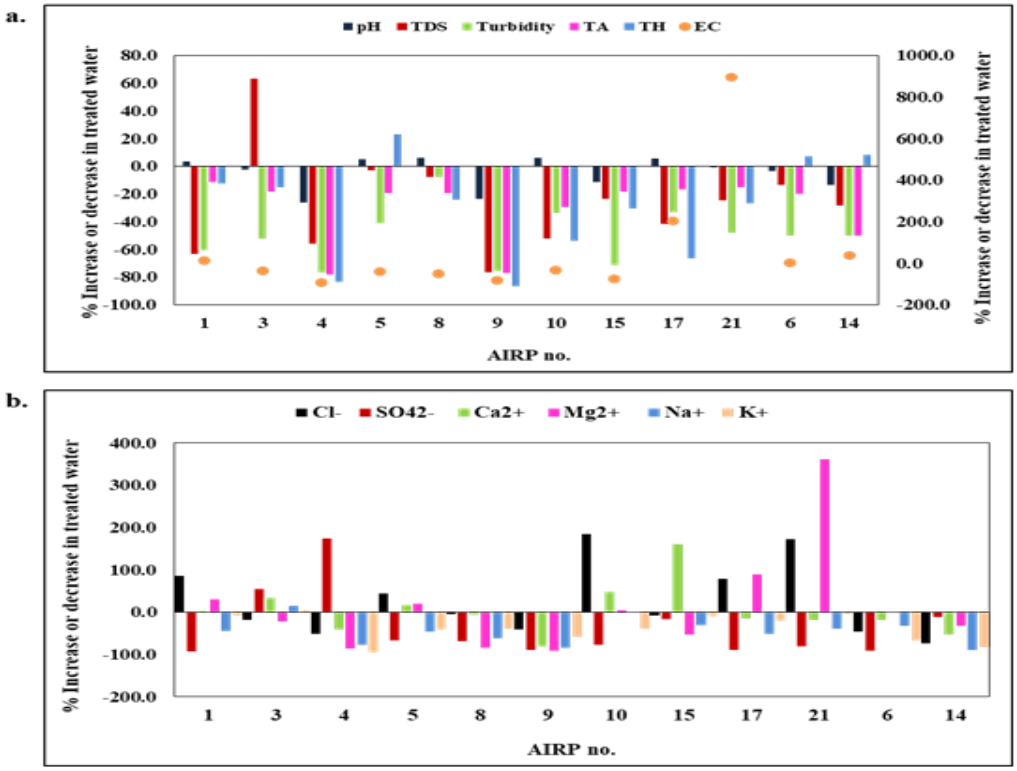


Fig. 59. Percentage of increase or decrease in the physicochemical parameters in the treated water from raw water

The treated water pH range lies between 5.79 and 7.83 for the 12 studied ATUs (**Table 50**) which is within the acceptable range of drinking water. The TDS in treated water of ARP 3 has purportedly been increased from raw water by almost 63.3% while that has been decreased in rest of the ARPs (range: 3 to 76.5%). The average TDS and TH concentrations of the studied plants (401 mg/L, 193 mg/L) are within the allowance of drinking water while that of TA (315 mg/L) is above the recommendation. It suggests that the drinking water quality is slightly alkaline in nature and the plants need to reduce the alkalinity to reach the recommendations. Electrical conductivity in treated water of the plants has been increased by an average of 61.5% (2208 µS/cm). EC has been radically increased by almost 205 and 894% in plant no. 17 and 21 while it has been decreased from raw water by nearly 93.3, 83.7 and 77.1% in plant no. 4, 9 and 15 respectively. Electrical conductanceis an indirect measure of the presence of dissolvedsolids in water samples (Hem, 1985). The high EC values above 1200µs/cmare the direct indication of the increasing mobility of ions in drinking watersources. Turbidity has been reduced in treated water for all the 12 selected plants by a range of 7.8 to 76.3% (Fig. 59a). The average turbidity in treated water of the studied plants is 7.96 NTU (range: 4.3 – 13 NTU), which

is higher than the recommendation. Total alkalinity (TA) is decreased in the treated water of all the selected ARPs by a range of 11.4 to 78%. The average TA is decreased from raw water (447 mg/L) to treated water (315 mg/L) by approximately 31.1 %. The average total hardness (TH) value in treated water of the plants is 193 mg/L (range: 53-333 mg/L). The TH in treated water is increased by 7.1 and 8.3 % in the plants no. 6 and 14 respectively, while in the remaining selected 10 plants, the TH is decreased by a range of 12.4 to 86.4%. The ionic concentration difference percentage between raw and treated water of the studied ARPs has been shown in Fig. 59**.** The average concentration of Cl^-, SO_4^{2-}, Ca^{2+}, Mg^{2+}, Na^+ and K^+ in treated water are 39.6, 1.98, 46, 47.8, 22.2 and 2.7 mg/L respectively. Annual average concentration of Cl^-in treated water has been increased in plants no. 1, 5, 10, 17 and 21 by 86.5, 44.6, 184, 79.8 and 173 %, possibly due to added chlorine during disinfection. Concentration of SO_4^{2-} is increased in treated water of plant no. 3 and 4 by 55 and 175%, however in the rest of the studied plants, it is decreased in a range of 10.9 to 93.3% (average: 37.7%). In the treated water samples, calcium concentration has been increased in the plants no. 1, 3, 5, 10 and 15 by 2.3, 34.2, 16.6 and 160 %, magnesium concentration has been enhanced in the plants no. 1, 5, 10, 17 and 21 by 29.6, 20.5, 3.8, 89.7 and 361%. Sodium concentration has decreased in almost all the ARPs excluding plant no. 3 and 10 while potassium concentration has declined in all the plants except ARP no. 3. Cumulatively, average sodium and potassium concentration in treated water is reduced from raw water by 44.9% and 38.1% (Fig. 59b). The average concentrations of chloride, sulphate, calcium and sodium in treated water samples are within the acceptable range while average magnesium concentration has crossed the required limit of 30 mg/L.

Table 50: Physicochemical parameters of treated water of the selected ARPs (Part A)

ARP no.	Parameters (analysed in three intervals round the year)																											
	pH				EC (µS/cm)				Turbidity (NTU)				TDS (mg/L)				TSS (mg/L)				Total Hardness (mg/L)				Total Alkalinity (mg/L)			
	1st	2nd	3rd	Avg.	1st	2nd	3rd	Avg.	1st	2nd	3rd	Avg.	1st	2nd	3rd	Avg.	1st	2nd	3rd	Avg.	1st	2nd	3rd	Avg.	1st	2nd	3rd	Avg.
1	7.86	7.22	8.04	7.71± 0.43	2150	2380	4160	2897 ± 1100	4	8	2	4.7 ± 3.1	750	750	400	633 ± 202	200	350	400	317 ± 104	300	320	380	333 ± 42	360	350	460	390 ± 61
3	7.91	7.52	7.65	7.69± 0.20	3680	4040	8200	5307 ± 2512	2	16	2	6.7 ± 8.1	850	700	733	761 ± 79	200	250	1150	533 ± 535	300	350	320	323 ± 25	430	450	420	433 ± 15
4	5.99	5.15	6.24	5.79± 0.57	370	258	545	391 ± 145	10	10	5	8.3 ± 2.9	300	350	166	272 ± 95	120	100	80	100 ± 20	40	60	70	57 ± 15	60	100	110	90 ± 26
5	7.83	7.26	8.18	7.76± 0.46	2160	2410	4450	3007 ± 1256	12	2	2	5.3 ± 5.8	500	550	500	517 ± 29	280	300	1250	610 ± 554	300	400	260	320 ± 72	400	420	340	387 ± 42
8	8.04	7.35	8.1	7.83± 0.42	2300	2440	2850	2530 ± 286	11	11	3	8.3 ± 4.6	500	520	366	462 ± 84	220	180	250	217 ± 35	260	250	330	280 ± 44	340	350	420	370 ± 44
9	6.17	5.04	6.21	5.81± 0.66	349	311	512	391 ± 107	10	10	2	7.3 ± 4.6	50	100	133	94 ± 42	160	120	600	293 ± 266	50	60	50	53 ± 6	80	80	170	110 ± 52
10	8.02	7.55	7.92	7.83± 0.25	2230	2380	5030	3213 ± 1575	17	12	5	11.3 ± 6.0	250	300	266	272 ± 26	240	250	100	197 ± 84	40	50	340	143 ± 170	390	350	400	380 ± 26
15	8.06	5.22	6.4	6.56± 1.43	3050	356	673	1360 ± 1472	3	7	3	4.3 ± 2.3	400	350	166	305 ± 123	340	280	500	373 ± 114	240	250	200	230 ± 26	510	500	120	377 ± 222
17	8.21	7.3	8.28	7.93± 0.55	2020	2330	2690	2347 ± 335	8	14	2	8.0 ± 6.0	220	350	600	390 ± 193	150	200	850	400 ± 391	90	100	130	107 ± 21	370	350	380	367 ± 15
21	7.89	7.52	7.94	7.78± 0.23	1650	1850	1750	1750± 100	11	10	4	8.3 ± 3.8	550	400	333	428 ± 111	180	200	750	377 ± 323	200	200	150	183 ± 29	320	300	350	323 ± 25
6	7.96	7.5	NA	7.73± 0.33	3040	3000	NA	3020 ± 28	16	10	NA	13.0 ± 4.2	500	450	NA	475 ± 35	280	250	NA	265 ± 21	230	220	NA	225 ± 7	510	450	NA	480 ± 42
14	6	7	NA	6.50± 0.71	249	300	NA	275 ± 36	10	10	NA	10.0 ± 0.0	250	150	NA	200 ± 71	200	250	NA	225 ± 35	50	80	NA	65 ± 21	60	90	NA	75 ± 21
Avg	**7.24 ± 0.23**				**2208 ± 1470**				**7.96 ± 2.60**				**401 ± 187**				**325 ± 144**				**193 ± 107**				**315 ± 140**			

Table 51: Physicochemical parameters of treated water of the selected ARPs (Part B)

ARP no.	Parameters (analysed thrice per year)																							
	Cl^- (mg/L)				SO_4^{2-} (mg/L)				Ca^{2+} (mg/L)				Mg^{2+} (mg/L)				Na^+ (mg/L)				K^+ (mg/L)			
	1st	2nd	3rd	Avg	1st	2nd	3rd	Avg	1st	2nd	3rd	Avg	1st	2nd	3rd	Avg	1st	2nd	3rd	Avg	1st	2nd	3rd	Avg
1	24.9	53.3	21.3	33.2 ± 17.5	0.1	0.1	0.1	0.1 ± 0.0	100	40	32	57.3 ± 37.2	84	70	73	75.7 ± 7.4	5.1	5.5	16.3	8.97 ± 6.35	2.7	3	3.1	2.93 ± 0.00
3	117	49.7	121	95.9 ± 40.1	5.93	3.28	0.2	3.1 ± 2.9	68	45	32	48.3 ± 18.2	52	54	58.4	54.8 ± 3.3	180	120	125	142 ± 33.3	3.6	5.1	4.1	4.27 ± 0.76
4	17.8	28.4	17.8	21.3 ± 6.1	3.82	5.68	0.3	3.3 ± 2.7	16	28	20	21.3 ± 6.1	5.2	15	4.87	8.4 ± 5.8	4.9	10	6.6	7.17 ± 2.6	1.5	1	0.5	1.00 ± 0.50
5	32	39.1	21.3	30.8 ± 9.0	1.08	0.1	0.1	0.4 ± 0.6	76	68	52	65.3 ± 12.2	42	32	31.6	35.2 ± 5.9	4.9	7	19.6	10.5 ± 7.95	1.8	1	3.5	2.10 ± 1.28
8	21.3	28.4	21.3	23.7 ± 4.1	7	2	5	4.7 ± 2.5	72	40	36	49.3 ± 19.7	10	10	8.4	9.5 ± 0.9	2.5	3	19.5	8.33 ± 9.67	1.7	1.5	3.4	2.20 ± 1.04
9	14.2	49.7	17.8	27.2 ± 19.5	0.1	0.1	0.1	0.1 ± 0.0	12	10	12	11.3 ± 1.2	3.5	6.8	4.87	5.1 ± 1.7	4.8	3.4	6.7	4.97 ± 1.66	1.5	1.8	0.5	1.27 ± 0.68
10	17.8	146	17.8	60.5 ± 74.0	4.31	0.1	2.5	2.3 ± 2.1	68	20	36	41.3 ± 24.4	71	50	60.8	60.6 ± 10.5	14.8	25	19.3	19.7 ± 5.11	1.6	1.5	3.5	2.20 ± 1.13
15	24.9	74.6	17.8	39.1 ± 30.9	5.39	4.21	3	4.2 ± 1.2	80	40	36	52.0 ± 24.3	45	25	26.8	32.3 ± 11.1	25.7	30	6.5	20.73 ± 12.5	4.1	3.5	0.5	2.70 ± 1.93
17	24.9	46.2	24.9	32.0 ± 12.3	0.1	0.1	0.1	0.1 ± 0.0	68	50	68	62.0 ± 10.4	76	55	63.2	64.7 ± 10.6	3.5	5	16.1	8.20 ± 6.88	3.6	3.6	4.1	3.77 ± 0.29
21	21.3	103	21.3	48.5 ± 47.2	0.1	0.5	0.1	0.2 ± 0.2	52	40	56	49.3 ± 8.3	96	115	124	112 ± 14.3	3.8	5	11.1	6.63 ± 3.91	2.5	2.8	3	2.77 ± 0.25
6	35.5	38	NA	36.8 ± 1.8	0.1	0.5	NA	0.3 ± 0.3	100	32	NA	66.0 ± 48.1	78	58	NA	68.0 ± 14.1	25.8	22	NA	23.90 ± 2.69	5.1	4.9	NA	5.00 ± 0.14
14	28.4	25	NA	26.7 ± 2.4	6.47	3.28	NA	4.9 ± 2.3	16	40	NA	28.0 ± 17.0	35	60	NA	47.5 ± 17. 7	4.8	5	NA	4.90 ± 2.3	1.4	3	NA	2.20 ± 1.13
Avg.	**39.6 ± 20.8**				**1.98 ± 1.98**				**46 ± 17.5**				**47.8 ± 31.7**				**22.2 ± 38.3**				**2.7 ± 1.17**			

5.6.2.5. Status of the four major toxic elements in raw and treated water of the selected ARPs

Concentration of As and other three major drinking water-pollutants (Fe, F^- and NO_3^-) in raw water and treated water (collected during three intervals in a year) is intricately given in **Table 52.** 11 out of the 12 selected ARPs in the present study used raw groundwater samples that contained much higher As concentrations. Arsenic concentration in the raw water sample was highest in ARP no. 6 (328 µg/L) and in the borderline of safe limit in ARP no. 3 (10 µg/L) respectively. The present study shows the extent of As contamination in groundwater samples with an average concentration of 154 µg/L (range: 10-328 µg/L) in the 12 selected plants' raw water. Groundwater As concentration range in several sites of North 24 Parganas district are earlier reported as 433 - 966 µg/L (average: 615 µg/L) in Mathpara, 29-829 µg/L (average: 301 µg/L) in Eithbhata, 3–169µg/L (average: 48 µg/L) in Madhusudankati and 8.5-55.8µg/L in Jamdani villages respectively (Joardar et al., 2021a). It was noticed that theaverage As concentration in treated water made by the ARPs were mostly unsafe for drinking. Annual average As concentration in treated water was almost 10 times higher in three ARPs; no. 6 (178 ± 19µg/L), no. 17 (102 ± 27µg/L) and no. 15 (96 ± 47.4µg/L) respectively. The annual average As concentration in treated water was in safe range in the ARP no. 3 (5 ± 2.7µg/L) as it used surface water as a source of raw water. Iron concentration range in the raw water of the ARPs were 2.1 mg/L (plant no. 8) to 10.07 mg/L (plant no. 17) and the average concentration was 4.97 mg/L. Similar Fe concentration range (0-15.29 mg/L) was observed by Singh et al. (2016) in the study area with an average concentration of 2.15 mg/L. The annual average iron concentration was found to be higher than the recommended value of drinking water (0.3 mg/L, WHO 2011) in the treated water supplied by ARP no. 5 (0.48 ± 0.37 mg/L), 10 (0.44 ± 0.26 mg/L), 15 (1.18 ± 1.82 mg/L), 17 (1.17 ± 1.46 mg/L), 21 (0.61 ± 0.22 mg/L) and 6 (1.53 ± 1.03 mg/L) respectively. Cumulative average Fe concentration of the treated water of the 12 plants (0.57 mg/L) is apparently 1.9 times higher than the recommendation of drinking water. Fluoride concentration was observed to be lower than the recommendation in each of the raw and treated water samples of the ARPs. The average F^- concentration in the raw and treated water of the 12 studied plants is 0.19 and 0.15 mg/L respectively (range: 0.15-3 mg/L and 0.03-0.29 mg/L). Average NO_3^-concentration in raw and treated water of the 12 ARPs was found to be 5.17 and 5.02 mg/L (range: 1.5-10.7 mg/L and 0.11-10.11 mg/L), much lower than the permissible limit of drinking water.

Table 52: Analysis of four major toxic elements in raw and treated water of the selected ARPs

ARP no.	Parameters																			
	Arsenic (µg/L)					Iron (mg/L)					Fluoride (mg/L)					Nitrate (mg/L)				
	Raw	Treated				Raw	Treated				Raw	Treated				Raw	Treated			
		1st	2nd	3rd	Avg.		1st	2nd	3rd	Avg.		1st	2nd	3rd	Avg.		1st	2nd	3rd	Avg.
1	153	10	55.4	36	34 ± 22.8	6.36	0.11	0.05	0.42	0.19 ± 0.2	0.15	0.12	0.11	0.17	0.13 ± 0.032	4.42	1.42	6.07	6.85	4.78 ± 2.93
3	10	7.74	3	3	5 ± 2.7	3.34	0.05	0.05	0.62	0.24 ± 0.33	0.3	0.23	0.32	0.31	0.29 ± 0.049	3.23	10.2	10.7	9.42	10.11 ± 0.65
4	280	<3	102	41	49 ± 49.9	6.21	0.05	0.05	0.4	0.17 ± 0.2	0.15	0.011	0.019	0.058	0.03 ± 0.025	6.19	0.13	0.56	4.71	1.80 ± 2.53
5	55.6	70	18.6	15	35 ± 30.8	4.28	0.16	0.39	0.89	0.48 ± 0.37	0.24	0.023	0.021	0.22	0.09 ± 0.114	5.75	0.044	6.42	4.71	3.72 ± 3.3
8	60.4	21.6	22.9	16	20 ± 3.7	2.1	0.05	0.12	0.72	0.30 ± 0.37	0.19	0.13	0.18	0.17	0.16 ± 0.026	6.63	1.01	10.7	9.84	7.18 ± 5.36
9	237	38.6	105	42	62 ± 37.4	7.47	0.16	0.05	0.35	0.19 ± 0.15	0.15	0.054	0.024	0.67	0.25 ± 0.365	3.40	1.05	7.7	3.38	4.04 ± 3.37
10	60.4	18.5	20.6	15	18 ± 2.8	2.5	0.14	0.57	0.62	0.44 ± 0.26	0.17	0.16	0.25	0.2	0.20 ± 0.045	10.7	15	6.42	2.91	8.11 ± 6.21
15	212	120	126	41	96 ± 47.4	2.77	3.28	0.21	0.05	1.18 ± 1.82	0.17	0.16	0.19	0.054	0.13 ± 0.071	4.11	7.08	4.62	6.42	6.04 ± 1.27
17	157	108	72.2	125	102 ± 27	10.07	0.63	0.05	2.82	1.17 ± 1.46	0.18	0.13	0.11	0.14	0.13 ± 0.015	4.29	6.2	1.5	4.28	3.99 ± 2.36
21	142	65.2	76.4	50	64 ± 13.3	6.76	0.47	0.5	0.87	0.61 ± 0.22	0.16	0.11	0.19	0.15	0.15 ± 0.040	5.30	4.87	1.2	3.30	3.12 ± 1.84
6	328	191	164	NA	178 ± 19	3.28	2.25	0.8	NA	1.53 ± 1.03	0.19	0.12	0.14	NA	0.13 ± 0.014	1.5	0.01	0.214	NA	0.11 ± 0.14
14	152	32.9	29.2	NA	31 ± 2.6	4.5	0.08	0.5	NA	0.29 ± 0.30	0.23	0.14	0.13	NA	0.14 ± 0.007	6.5	3.85	10.7	NA	7.28 ± 4.84
Avg.	154	57.2	66.3	38.4	57.8 ± 48.2	4.97	0.62	0.28	0.78	0.57 ± 0.47	0.19	0.12	0.14	0.21	0.15 ± 0.07	5.17	4.24	5.57	5.58	5.02 ± 2.83
	Permissible limit in drinking water				10	**Permissible limit in drinking water**				0.3	**Permissible limit in drinking water**				1.5	**Permissible limit in drinking water**				45

5.6.2.6. Arsenic and iron removal efficiency of the ARPs

During the annual sample collection in a year, the average As removal efficiency of the ARPs were observed in a range between 35.2 to 82.6 % (average: 61.2 %). Average As removal efficiency of the studied units during three intervals of sampling observed as 54.6 % (range: -25.9 to 98.9 %), 59.9 % (range: 40.6 to 80.8 %) and 70.2 % (range: 20.4 to 85.4 %). At the 1st time of sampling, ARP no. 5 showed -25.9 % As removal efficiency, which means that the treated water contained As concentration more than raw water. It may be caused by the backflow of As through the membranes. ARP no. 17 showed the poorest performance throughout the year; As removal percentage was seen to be 31.2 %, 54 % and 20.4 % during the three respective sampling time. ARP no. 6 was also not much efficient in removing As as the removal was observed to be 41.8 % and 50 % during the 1st and 2nd sampling. Moreover, due to ill maintenance, ARP no. 6 and 14 got closed and 3rd time sampling was not possible. This distressing picture is common in most of the As endemic sites as Hossain et al. (2006) too showed that 4plants out of total studied 18 ARPs in Baruipur block of the South 24-Parganas district were officially abandoned by the project authority because of poor efficiency and choking the tube well and the filter media known as sand gushing. In the present study, ARP no. 3 is actually a surface water treatment plant as it uses pond water for treatment (As concentration 10 µg/L). During the three time of sampling, it is observed that the plant produces As-safe water with an average As removal efficiency of 50 %. The average As removal efficiency of the 12 ARPs along with their efficiency at three respective time of sampling in the whole year is shown in **Fig. 60**.

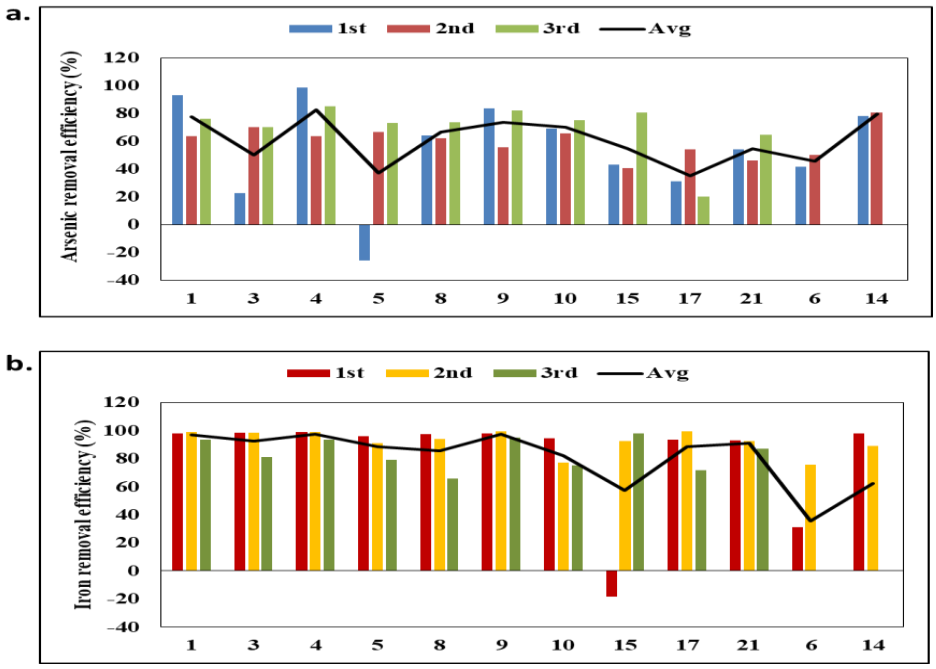


Fig. 60. Arsenic and iron removal percentage of the ARPs

It is surprising to see that most of the ARPs (5, 8, 9, 10, 15, 17, 21 6, 14) failed to reach the safe limit of As concentration in drinking water (suggested by WHO) during the three time of sampling. In some instances, the proficiency of the ARPs in removal of As decreases with time and during the survey, the information is obtained that the membranes or filters are not cleaned or changed regularly. It reveals that although the ARPs remove As substantially, the effectiveness is not maintained throughout the year which is caused by lack of monitoring and ample negligence. Sorenson and Mcbean (2015) reported that on an average, at around 8days after cleaning the arsenic removal efficiency of the ARPs is maximum, and then it declines until the next cleaning. Rahman et al. (2021) also reported that As removal efficiency of 20 studied ARPs in Southwest Bangladesh ranged between 67% - 98% and 74 - 93% during pre and post monsoon periods respectively. Basak et al. (2021) performed a critical review on the performance of four different technologies of ARPs in North 24 Pargana and showed that how the nuisances like irregular backwashing of filters, poor sludge treatment, inadequate chemical dose, damage of pipe and pump due to chemical corrosion, poor or no washing of adsorption bed can cause sudden increase of As in treated water. To avoid blockage of the media and reinstate its finest water treatment efficacy, the system needs periodical cleaning by backwashing that also produces huge amount of wastewater (Koley, 2021). Similarly, Sorensen and McBean (2015) reported that in rural Bangladesh, the efficiency of ARPs declined by 10% after 3 years of installation due to interrupted use and improper preservation; however, the initial performance of ARPs was satisfactory. On the other hand, it is seen that except plant no. 6 and 15, the rest 10 ARPs are potent to remove iron at a significant level. The average iron removal percentage of the plants is 81.4% (range: 35.7% to 97.3%). ARP no. 15 shows iron removal percentage of -18.4, 92.4 and 98.2 at the time of 1^{st}, 2^{nd} and 3^{rd} sampling while ARP no. 6 shows 31.4% and 75.6% iron removal efficiency at the two respective sampling time. The annual average iron removal efficiency of these two ARPs is 57.4 % and 35.7 % respectively. Average iron removal efficacy of the studied units during three intervals of sampling observed as 81.7% (range: -18.4 to 99%), 92.3% (range: 75.6 to 99.5%) and 70% (range: 35.6 to 97.5%). The iron removal efficiency of the 12 ARPs at three consecutive time of sampling in the whole year as well as their average efficiency percentage is shown in Fig. 60b.

5.6.2.7. Fe/As molar ratio

Studies reveal that naturally occurring Fe content in groundwater plays a crucial role in As removal (Berg et al., 2006; Mamtaz et al., 2001; Sharma et al., 2016). Presence of Fe with As in water improved the As deduction through co-precipitation (Bhavan, 2007). Therefore, Fe/As ratio is important in the iron-based adsorbent technologies for arsenic removal. Reports say that average molar Fe/As ratio in the groundwater of Vietnam was between 60 and 68 (Berg et al., 2008) while that in Raninagar-II, Murshidabad lies between 19 and 33.2. We found that dug-wells seem to be the safest source of drinking water having Fe/As value of 66.1 (range: 9-122). The Fe/As value in raw water of Sajaldhara treatment plant and Reverse Osmosis plants were 66.9 and 52.2 respectively while that in treated water were 146 and 15.3. Higher Fe/As value is required to attain As-safe drinking water (Banerji and Chaudhari, 2017; Schmidt et al., 2016). In a report of Vietnam, it was declared that the filtering system (sand filtration) can trim down the As levels below the permissible limit when the Fe/As ratio was ≥ 250 (Berg et al., 2006). The present study observed average Fe/As ratio of 62.3 (range: 10- 334) and 13.7 (range: 3.06 - 48) in the raw and treated water of the ARPs respectively. The raw and treated water Fe/As molar ratio of the 12 ARPs are shown in **Fig. 61a**.

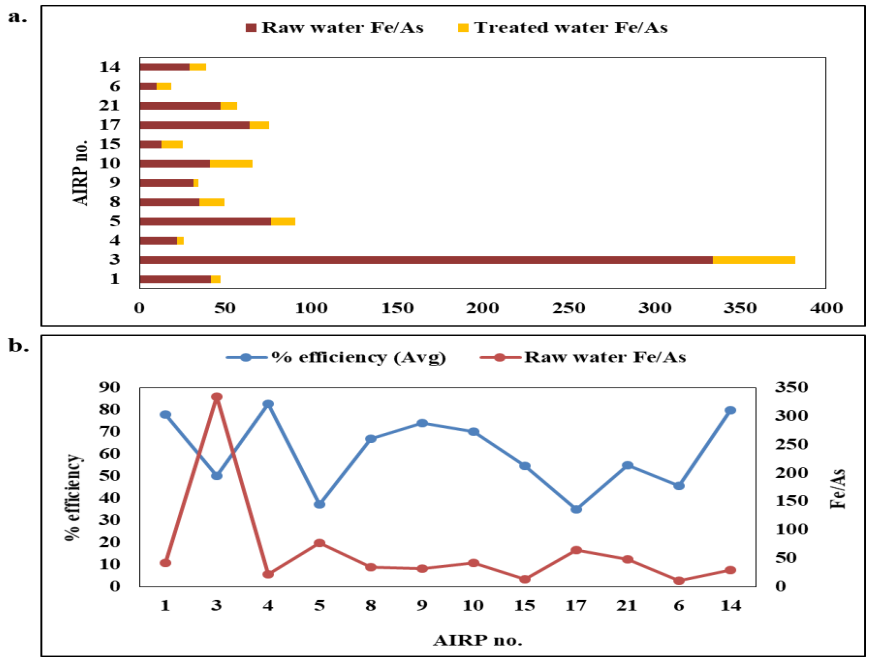


Fig. 61. Fe/As molar ratio of the ARPs

The surface water treatment plant showed highest Fe/As value (334 and 48) in its raw and treated water and its removal efficiency is seen to be 50%. The removal efficiency and

Fe/As ratio is compared in **Fig. 61b**. The regression statistics for the 11 ARPs (excluding the surface water treatment plant) reveals that the R^2 value is 0.65 ($p< 0.05$) between their Fe/As ratio and As removal efficiency which signifies a moderate association. Similar kind of significant correlation between Fe/As ratio and As removal efficiency were observed in treated water supplied by the plants in pre and post monsoon in rural southwest Bangladesh (Rahman et al., 2021). Fe/As ratio of 30 was observed to attain a As removal efficiency of 75% in the community-based ARPs in Bangladesh. Whereas, to achieve 80-90 % As removal efficiencies, the United States Environmental Protection Agency endorses the Fe/As ratio around 20 (Ahmad, 2005 and USEPA, 2005). Therefore, it is important to look into the function of Fe/As ratio in the As removal capacities of ARPs.

5.6.2.8. Correlation among the physicochemical parameters

The correlation coefficient value (r) between two parameters describes the degree of association between them. **Fig. 62** shows the extent of inter-relation among the physicochemical parameters analyzed for treated water quality evaluation of the ARPs. The degree of positive correlation is classified into five categories and designated with five different colors whereas the negative associations are highlighted in a yellow box. In the present study, pH seemed to be very strongly correlated with EC (r = 0.805) and calcium ions (r = 0.831), while, EC is also very strongly associated with TDS (r = 0.859) and TA (r = 0.839). The strong relation of TA with TDS (r = 0.727) and pH (r = 0.839) along with the average TA value (Table 3A) suggests that the treated water quality tends to be alkaline in nature. Measurement of conductivity actually determines the total number of ions present in a water sample, therefore, higher the TDS value, higher the conductivity. Thus, the ionic conductance or EC is strongly correlated with Ca^{2+} ions (r = 0.650), Na^{+} ions (r = 0.707), K^{+} ions (r = 0.668), Cl^{-} ions (r = 0.769), moderately associated with Mg^{2+} ions (r = 0.439) and F^{-} ions (r = 0.435) and negatively allied with SO_4^{2-}ions (r = -0.198). We found similar kind of strong positive relation of EC with TDS (r = 1) and moderate positive association of EC with TA and TDS (r = 0.48 and 0.48) respectively in groundwater samples of Nadia district. In a same way, TDS in treated water samples is moderately linked with Cl^{-} (r = 0.537) and Mg^{2+} (r = 0.405), strongly correlated with Ca^{2+}(r = 0.688), Na^{+}(r = 0.621), K^{+} (r = 0.626) and negatively associated with SO_4^{2-}ions (r = -0.157). Quite strong correlation between TDS and TH (r = 0.867) signifies that the minerals responsible for TDS value in drinking water causes hardness of water. Both hardness and alkalinity of the treated water samples are strongly linked to each other (r =

0.782) and mostly regulated by calcium ions compared to magnesium ions, which are well demonstrated by their corresponding r values. A strong positive correlation exists between TH and Ca^{2+} ions (r = 0.716), while a weak correlation is found between TH and Mg^{2+} ions (r = 0.23). Sodium and potassium ions concentrations are moderately related with hardness of water (r = 0.424 and 0.448). Alkalinity is determined by a very strong association with Ca^{2+}, strong association with K^{+} ions, moderate association with $Mg^{2+}$and Fe ions and weak association with Na+ ions (r = 0.885, 0.739, 0.455, 0.511 and 0.369 respectively). The association between the anions (sulphate and chloride) and alkalinity of water are observed negative (r = -0.268) to moderately positive (r = 0.465). A strong and significant positive association in treated water of the plants is observed between arsenic and iron (r = 0.864) which corroborates with the importance of Fe/As molar ratio. We also reported the interrelation of groundwater As and iron concentration in agricultural shallow tube-wells (R^2 = 0.816) and domestic and community level shallow tube-wells (R^2 = 0.515). Arsenic concentration in treated water samples of these selected plants is found to be insignificantly or negatively related with most of the ions; F^{-} (r = -0.306), Cl^{-} (r = -0.323), Na^{+}(r = -0.328), NO_3^{-} (r = - 0.705) and SO_4^{2-} (r = -0.413). NO_3^{-} in these water samples are moderate to strongly connected with Cl^{-} (r = 0.579), SO_4^{2-}(r = 0.570), Na^{+} (r = 0.546) and F^{-}(r = 0.625) while conversely related to Ca^{2+}(r = - 0.120), Mg^{2+}(r =- 0.085), K^{+}(r = - 0.019) and Fe (r = - 0.426). Fe concentration in water samples is also negatively associated with Cl^{-} (r = -0.047), SO_4^{2-} (r = -0.235) or F^{-} ions (r = -0.2). These kind of interrelations among the cations and anions in groundwater are also found in several other studies of India (Achary et al., 2014, Ramakrishnaiah et al., 2009; Dev and Bali, 2019).

	pH	EC	TDS	TSS	TH	TA	Cl-	SO42-	Ca2+	Mg2+	Na+	K+	As	F	Fe	NO3-
pH	1.000															
EC	0.805	1														
TDS	0.690	0.859	1													
TSS	0.428	0.557	0.562	1												
TH	0.643	0.756	0.867	0.586	1											
TA	0.839	0.839	0.727	0.490	0.782	1										
Cl-	0.384	0.769	0.537	0.428	0.374	0.465	1									
SO42-	-0.299	-0.198	-0.157	-0.348	-0.077	-0.268	0.035	1								
Ca2+	0.831	0.650	0.688	0.542	0.716	0.885	0.174	-0.309	1							
Mg2+	0.623	0.439	0.405	0.260	0.230	0.455	0.393	-0.452	0.517	1						
Na+	0.215	0.707	0.621	0.447	0.424	0.369	0.886	0.179	0.122	0.096	1					
K+	0.629	0.668	0.626	0.376	0.448	0.739	0.489	-0.246	0.707	0.578	0.502	1				
As	-0.060	-0.241	-0.220	-0.015	-0.219	0.182	-0.323	-0.413	0.298	0.167	-0.328	0.376	1			
F	0.140	0.435	0.144	0.250	0.136	0.183	0.683	-0.013	-0.178	0.064	0.615	0.246	-0.306	1		
Fe	0.288	0.100	0.016	0.142	0.039	0.511	-0.047	-0.235	0.599	0.322	-0.103	0.646	0.864	-0.200	1	
NO3-	0.162	0.358	0.192	0.169	0.230	0.086	0.579	0.570	-0.120	-0.085	0.546	-0.019	-0.705	0.625	-0.426	1

Positive association

- Very strong association (r = 0.8-1.0)
- Strong association (r = 0.6-0.79)
- Moderate association (r = 0.4-0.59)
- Weak association (r = 0.2-0.39)
- Very weak association (r = 0.00-0.19)

*Yellow highlighted boxes are negative association

Fig. 62. Pearson correlation among the parameters in treated water

5.6.2.9. Hierarchical cluster analysis among the physico-chemical parameters

A hierarchical cluster analysis helps to plot a dendrogram through a single linkage algorithm method maintaining a Euclidean distance between two points of the data. In the present study, the relationship among the parameters analyzed for raw and treated water samples are shown in **Fig. 63**.

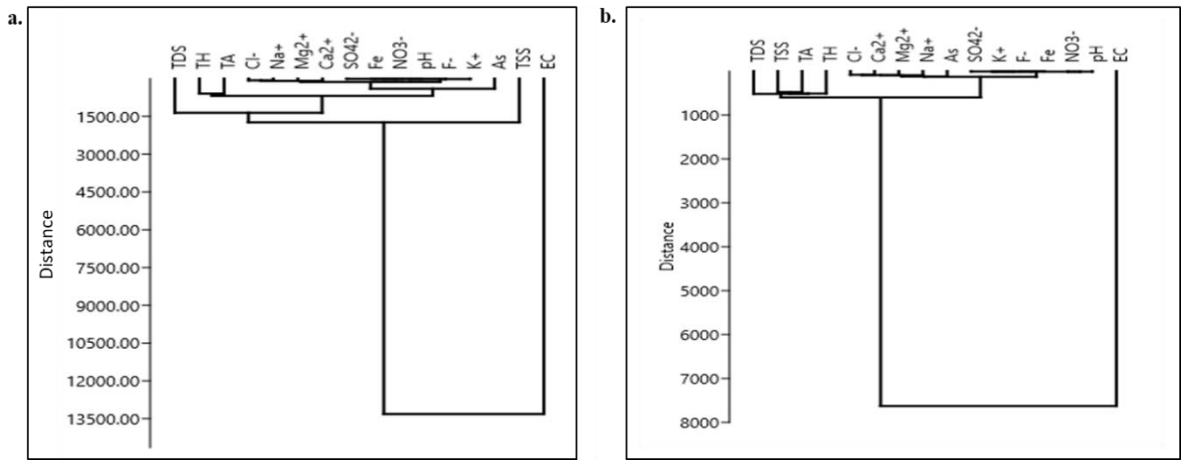


Fig. 63. Hierarchical cluster analysis among the parameters a) Raw water b) Treated water

It is seen that there is a direct relationship between total hardness (TH) and total alkalinity (TA) in both raw and treated water samples (Fig. 68a, 68b). The ions in raw groundwater samples show their strong inter-personal association through different short and single clusters (Cl-Na^{+}, Na^{+}-Mg^{2+}, Mg^{2+}-Ca^{2+}, Ca^{2+}-SO_4^{2-} etc. It is also understood that distribution of all the ions is linked to TH, TDS (total dissolved solids) and TA of water. Total suspended solids (TSS) also make a good association with TDS as well as some other ions. **Fig. 63a** shows that As concentrations in raw groundwater samples are greatly related to iron concentrations. It is clearly seen that there is a direct relationship between TDS and TSS, TSS and TA, TA and TH, which again creates an association with the presence of the different ionic concentrations in the treated water samples (**Fig. 63b**). The most dominant and major cluster forms between electrical conductivity (EC) of the water samples with the ionic concentrations in water along with TDS, TSS, hardness and alkalinity in both the raw and treated water samples. Conductivity is originally attributed to dissolved ions and minerals in groundwater which affects water quality (Oparaocha et al., 2010).

5.6.2.10. Essential and non-essential multi-elements in drinking water of the selected ARPs

Apart from the general physicochemical parameters, it is important to determine the presences of essential and non-essential multi-elements in drinking water, which is given in **Table 53**.

Table 53: Concentration of trace elements and heavy metals in drinking water of the selected ARPs

ARPno.	Gram Panchayat	Type of water (Treated water of 3rd time collection)	Parameters								
			Al (µg/L)	Cr (µg/L)	Mn (µg/L)	Cu (µg/L)	Zn (µg/L)	Se (µg/L)	Cd (µg/L)	Pb (µg/L)	Hg (µg/L)
1	**Sutia**	**Raw**	52	0.30	184	1.85	12.8	0.061	0.024	2.66	<0.00
		Treated	26.6	0.16	1.4	1.86	6.29	0.152	0.013	0.84	0.002
3		**Raw**	9.89	0.267	90.6	2.60	2878	1.013	0.011	2.09	0.03
		Treated	***15.06***	0.107	***258***	0.81	18.4	0.502	0.007	0.57	0.026
4	**Ichapur I**	**Raw**	10.3	0.090	158	0.45	1.83	0.051	0.007	0.20	<0.00
		Treated	8.62	0.083	6.97	0.37	2.08	<0.000	0.013	0.29	<0.00
5		**Raw**	11.7	0.350	52.6	0.86	123	0.104	0.004	1.49	<0.00
		Treated	9.79	0.142	***79.2***	0.54	4.51	<0.000	0.004	0.14	<0.00
8	**Chandpara**	**Raw**	14.9	0.855	55.5	1.22	130	0.038	0.013	1.73	<0.00
		Treated	***15.9***	0.126	***82.9***	0.54	2.74	0.025	0.012	0.27	<0.00
9		**Raw**	8.00	0.199	157	0.73	1.78	0.124	0.003	0.24	<0.00
		Treated	9.80	0.090	6.91	15.7	***74.6***	<0.000	0.021	0.88	0.0006
10	**Dooma**	**Raw**	20.4	0.446	57	1.37	152	0.052	0.020	2.37	<0.00
		Treated	17.1	0.047	***81.7***	1.00	10.8	<0.000	0.010	0.27	<0.00
15	**Shimulpur**	**Raw**	9.39	0.085	163	0.58	1.69	0.086	0.007	0.13	0.002
		Treated	***11.6***	0.075	7.15	27.5	***4.56***	0.063	0.032	0.46	<0.00
17	**Jaleswar I**	**Raw**	17.9	0.382	105	1.10	911	0.129	0.037	4.68	<0.00
		Treated	14.5	0.277	***290***	4.68	22.6	0.014	0.010	0.49	<0.00
21	**Dharampur I**	**Raw**	16.5	0.113	76.7	1.79	10.9	0.084	0.008	0.28	<0.00
		Treated	12.9	0.059	52.8	0.675	4.83	0.017	0.007	0.35	<0.00
Acceptable limit of elements in drinking water (µg/L) (BIS, 2012**)**			30	50	100	50	5000	10	3	10	1
Permissible limit in absence of any alternative source (µg/L) (BIS, 2012)			200	No relaxation	300	1500	15000	No relaxation	No relaxation	No relaxation	No relaxation

Deficit of essential micronutrients in drinking water enhances the likelihood of suffering from As toxicity. Selenium (Se) and Zinc (Zn) are essential micronutrients in drinking water having and anti-toxic properties towards As (Otieno, 2017). Higher As concentrations replace Se in the Se dependent enzymes of mammalian body and inactivate it (Sun et al., 2014). Also, Zn increases the level of As detoxifying protein metallothionein (Roychowdhury et al., 2003). Both Se and Zn concentrations in the treated drinking water samples are very low and undeniably within their respective acceptable limits in drinking water. However, in the plants no. 15 and 9, Zn concentration is increased in treated water from their raw water samples by almost 41.9 and 2.7 times. Raw water sample of plant no. 3 and 17 showed substantial amount of Zn concentration (2878 and 911 µg/L), while that in the treated water sample was non-significant (18.4 and 22.6 µg/L) respectively. In another severe As contaminated block in North 24 Parganas district named Deganga, Rahman et al. (2015) reported similar trend of Se and Zn concentration in groundwater sample as <0.2-5.1 µg/L and 2-299 µg/L (n= 60). The ARPs are efficient to maintain the aluminium (Al) concentration within the acceptable range; however, the concentration is increased in treated water samples from raw water by approximately 52.3 %, 6.71 % and 23.5 % in plants no. 3, 8 and 15 respectively (Table 53). This may result from addition of Al based coagulants during treatment of water for removal of turbidity, organic matter and microorganisms. Being a non-essential element, higher Al concentrations in drinking water can accumulate in the human body and cause several health problems like Alzheimer's disease (He et al., 2021). Public health concerns are addressed on the occurrence of non-essential hexavalent chromium (Cr-VI) that is a known human carcinogen (Zhitkovich et al., 2011). In the present study, Cr concentration in drinking water is very low compared to its acceptable limit drinking water. Manganese (Mn), an essential trace element, acts as an important component of several enzymes and takes part in number of vital physiological processes in human body (WHO, 2021). In the present study, concentration of Mn is increased from raw water in treated water by 185, 50.6, 49.4, 43.3 and 176 % in the plants no. 3, 5, 8, 10 and 17 respectively. Significantly, Mn concentration is observed to be higher than its acceptable limit in drinking water in the treated water sample of ARP no. 3 (258 µg/L) and 17 (290 µg/L). This may be caused by the excessive use MnO_2 during oxidation of the pumped groundwater. Maximum ARPs OR ATUs have been seen to use MnO_2 during or prior sand filtration. Adverse health effects regarding memory, attention, and motor skills may occur in children and adults who consume excess Mn through drinking water. Infants may grow learning and behavior

glitches if they consume Mn contaminated water (WHO, 2021; Oulhote, 2014). Copper (Cu) is another essential trace element in drinking water, whose concentration is apparently low in the studied samples and certainly under the allowed limit in drinking water (50 µg/L). Cadmium (Cd), Lead (Pb) and mercury (Hg) are the non-essential trace elements in drinking water and their respective concentrations in treated drinking water samples of all the selected ATUs are below their acceptable limit in drinking water. It is also seen that As concentration is negatively correlated with Al (r = -0.32), Mn (r = -0.014), Se (r = -0.451), and Hg (r = -0.484). Poor/nil correlation between As and Mn was also observed in groundwater samples of Bengal delta as reported by Nath et al. (2011) and Rahman et al. (2015). Significant positive association is observed between As and Cu (r = 0.624) as well as Cd (r = 0.526) concentrations (**Table 54**).

Table 54. Correlation matrix among the concentration of multi-elements and arsenic in the treated water:

	As	Al	Cr	Mn	Cu	Zn	Se	Cd	Pb	Hg
As	1									
Al	-0.320	1.000								
Cr	0.339	0.236	1.000							
Mn	-0.014	0.043	0.601	1.000						
Cu	0.624	-0.275	-0.154	-0.301	1.000					
Zn	0.178	-0.232	0.049	0.022	0.351	1.000				
Se	-0.451	0.298	0.009	0.479	-0.128	-0.025	1.000			
Cd	0.526	-0.153	-0.231	-0.463	0.932	0.249	-0.179	1.000		
Pb	0.152	0.384	0.151	-0.097	0.359	0.662	0.307	0.374	1.000	
Hg	-0.484	0.134	-0.029	0.547	-0.181	0.063	0.974	-0.258	0.225	1

Comparable moderate positive associations between As and Cu and Cd were observed in groundwater samples of Pesarlanka Island, Krishna Delta, India (Mondal et al., 2010b). At the same time, a substantial correlation is found between Cu and Cd (r = 0.932), Se and Hg (r = 0.974), Pb and Zn (r = 0.662), Mn and Cr (r = 0.601) respectively. Mondal et al. (2010b) also reported similar kind of observations between Mn and Pb and Zn concentrations. The present study observed an insignificant but negative correlation between Se and Zn concentrations (r = -0.025), which is corroborated with findings of Alqahtani et al. (2021). On the other hand, **Fig. 64** shows the pattern of existence of As in water samples with other essential and non-essential elements through principal component analysis.

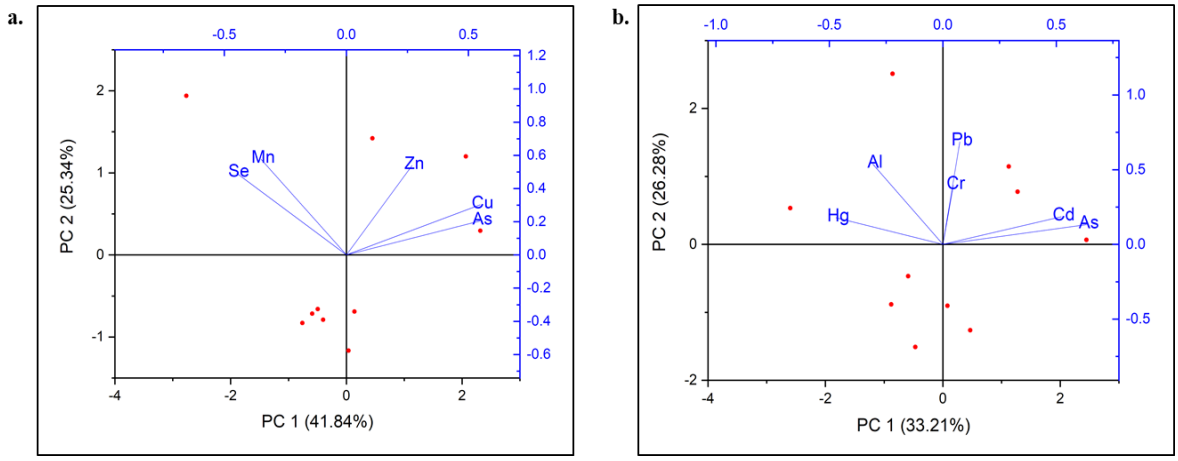


Fig. 64. Principal component analysis among multi-elements: a) arsenic and essential trace elements, b) arsenic and non-essential trace elements

It justifies the correlation values described before, i.e. Se and Mn are not controlled by As concentration in the samples, while Zn and Cu are linked with As and falls under same quadrant. The two principal components in essential trace elements, contribute by 41.84% and 25.34% respectively (Fig. 64a). Fig. 64b explains that the metalloid As and the heavy metals Cd, Cr and Pb play similar role in the water samples as they fall under the same quadrant while Al and Hg work differently. The eigen values and percentage of variance are shown elaborately in **Table 55**.

Table 55: Eigen values in Principal component analysis among arsenic and other multi-elements

	Essential trace elements		
	Eigen value	**Percentage of variance**	**Cumulative variance**
1	2.09203	41.84 %	41.84 %
2	1.26697	25.34 %	67.18 %
3	0.85084	17.02 %	84.20 %
4	0.67248	13.45 %	97.65 %
5	0.11768	2.35 %	100.00 %
	Non-essential trace elements		
1	1.99284	33.21 %	33.21 %
2	1.57651	26.28 %	59.49 %
3	1.22795	20.47 %	79.95 %
4	0.77186	12.86 %	92.82 %
5	0.27462	4.58 %	97.40 %
6	0.15622	2.60 %	100.00 %

Table 56: Extracted eigen vectors in Principal component analysis among arsenic and other multi-elements

	Coefficients of PC 1	Coefficients of PC 2
	Essential trace elements	
As	0.54833	0.20522
Zn	0.26924	0.5388
Se	-0.45021	0.49297
Mn	-0.35006	0.57693
Cu	0.54919	0.30283
	Non-essential trace elements	
As	0.63357	0.13262
Al	-0.30865	0.53782
Cr	0.04671	0.39458
Cd	0.52292	0.18496
Pb	0.0745	0.68648
Hg	-0.47133	0.17889

5.6.2.11. Indexing of treated water qualityof the ARPs (WQI, HEI)

5.6.2.11.2. Water quality index (WQI)

A water quality indexing (WQI) has been done considering 21 parameters (pH, TDS, Cl, SO_4^{2-}, TH, TA, Ca, Mg, Na, K, F^-, NO_3^-, Fe, As, Al, Cu, Mn, Cr, Cd, Pb, Hg) in treated drinking water samples of the selected ARPs. WQI proposes a scale reflecting a complete picture towards the water quality of the samples (Tiwari and Mishra, 1985). The parameters are assigned with a weight (wi) accordingto their importance in view of their contribution in the overall quality of waterfor drinking purpose. Parameters along with their allotted weights and calculated relative weights are given in **Table 57**.

Table 57: Relative weight of the parameters for WQI estimation

Parameters	Unit	Weight	Relative weight
pH	-	3.00	0.0441
TDS	mg/L	2.00	0.0294
Cl	mg/L	3.00	0.0441
SO4	mg/L	2.00	0.0294
TH	mg/L	2.00	0.0294
TA	mg/L	2.00	0.0294
Ca	mg/L	2.00	0.0294
Mg	mg/L	2.00	0.0294
Na	mg/L	2.00	0.0294
K	mg/L	2.00	0.0294
F	mg/L	4.00	0.0588
NO_3	mg/L	4.00	0.0588
Fe	mg/L	3.00	0.0441
As	µg/L	5.00	0.0735
Al	µg/L	2.00	0.0294
Cu	µg/L	4.00	0.0588
Mn	µg/L	4.00	0.0588
Cr	µg/L	5.00	0.0735
Cd	µg/L	5.00	0.0735
Pb	µg/L	5.00	0.0735
Hg	µg/L	5.00	0.0735
		$\sum = 68$	$\sum = 1.00$

The calculated Water Quality Index (WQI) values (through eq. 2 to 4) in the present work suggest that plant no. 15, 17 and 6 provide poor quality of drinking water (WQI = 117, 138 and 184 respectively). Drinking water supplied by the plant no. 14 can only be termed as ‘excellent’ having the lowest WQI value of 43.1 and the rest ARPs deliver ‘good’ quality of water. Estimated WQI values of the ARPs are given in Table 57 with their corresponding remarks. Remarkably, 25% treated water sample is observed with ‘poor’ quality.

5.6.2.11.3. Heavy metal evaluation index (HEI)

The calculated HEI values (eq. 8 to eq. 9) range from 3.04 to 17.11 and mean value is 7.92. Therefore, It is observed that plant no. 17 has the highest HEI value (17.1) indicating 'high' pollution followed by plant no. 6 with HEI value of 16.92. ARP no. 15 designates 'medium' category of pollution with HEI value of 14.2 and the rest 9 plants are of 'low' pollution. Notably, 16.7 % treated water sample is observed with 'high' heavy metal evaluation index value.

Table 58: Indexing of water quality (WQI, HEI)

ARP no.	WQI	Remarks	HEI	Remarks
1	63.2	Good	4.17	Low
3	60.0	Good	3.94	Low
4	50.5	Good	5.53	Low
5	65.3	Good	5.87	Low
8	49.0	Good	3.88	Low
9	62.4	Good	7.29	Low
10	52.5	Good	4.15	Low
15	117	Poor	14.2	Medium
17	138	Poor	17.1	High
21	91.5	Good	9.01	Low
6	184	Poor	16.92	High
14	43.1	Excellent	3.04	Low

5.6.2.12. Health risk assessment

Health risk assessment study (eq. 10 to eq. 12) shows that the population suffers from sufficient cancer and non-cancer risk (average: 2.77×10^{-3} and 6.17) from the treated water supplied arsenic concentration (**Table 59**).

Table 59: Health risk assessment

Parameters	Hg	Al	Cd	Cr	Pb	F	NO_3	As	
RfD value	0.0003	1.00	0.0005	0.003	0.0035	0.06	1.6	0.0003	
CSF	-	-	-	-	-	-	-	1.5 per mg kg/bw/day	
ARP no.	**Non cancer risk (HQ); Threshold value = 1**								**Cancer risk (CR); Threshold value = 10^{-6}**
1	0.24	0.032	0.31	0.04	0.86	0.05	0.0024	4.02	1.81×10^{-3}
3	3.10	0.018	0.17	0.03	0.58	0.11	0.0050	0.55	2.45×10^{-4}
4	-	0.010	0.31	0.02	0.30	0.01	0.0009	5.79	2.60×10^{-3}
5	-	0.012	0.10	0.03	0.14	0.03	0.0018	4.11	1.85×10^{-3}
8	-	0.019	0.29	0.03	0.28	0.06	0.0036	2.40	1.08×10^{-3}
9	-	0.012	0.50	0.02	0.90	0.10	0.0020	7.37	3.31×10^{-3}
10	-	0.020	0.24	0.01	0.28	0.08	0.0040	2.15	9.61×10^{-4}
15	-	0.014	0.76	0.02	0.47	0.05	0.0030	11.39	5.12×10^{-3}
17	-	0.017	0.24	0.07	0.50	0.05	0.0020	12.11	5.45×10^{-3}
21	-	0.015	0.17	0.01	0.36	0.06	0.0015	7.60	3.42×10^{-3}
6	-	-	-	-	-	0.05	0.0001	14.09	6.34×10^{-3}
14	-	-	-	-	-	0.05	0.0036	2.46	1.10×10^{-3}
Avg.	**0.67**	**0.01**	**0.26**	**0.02**	**0.39**	**0.06**	**0.0025**	**6.17**	**2.77×10^{-3}**

It is surprising to see that all the studied ARPs provide higher amount of cancer risk than the acceptable limit of USEPA (1×10^{-6}). The estimated cancer risk ranges between 2.45×10^{-4} (plant no. 3) and 6.33×10^{-3}(plant no. 6) with an average of 2.77×10^{-3}. On the other hand, except plant no. 3 (HQ_{As} = 0.55), rest of the 11 plants (91.7 %) provide higher amount of non-cancer risk than the threshold limit i.e. 1; the HQ_{As} values for plant no. 6, 17 and 15 are really alarming (14.09, 12.1, 11.4 respectively). The water samples are safe from the point of non-cancer risk through presence of other toxic elements; average HQ values for Hg, Al, Cd, Cr, Pb, F^-, $NO_3$are 0.67, 0.01, 0.26, 0.02, 0.39, 0.06 and 0.0025 respectively. But the plant no. 3 promotes a good amount of non-cancer risk through presence of Hg in the supplied water (HQ_{Hg} = 3.10). Bortey-Sam et al. (2015) stated that if HQ value is between 1 and 4, the health risk is in 'medium' order. Apart from As, Hg also effects human health adversely. Mercury can impair any organ and lead to malfunctioning of nerves, kidneys and muscles; human brain is the main target organ of it. It can also cause trouble to the membrane potential and interrupt with intracellular calcium homeostasis (Patrick, 2003). So, it is important to take care of the treated drinking water prior to reach the inhabitants.

5.6.3. Dug well

In cases, the As removal plants are run by some individuals and non-profit organizations, the drinking water is not provided at no cost to the local inhabitants. The poor economic status and incognizance force the inhabitants suffer more from water-borne diseases as they continue consuming As-contaminated water from the available domestic and community level shallow tube-wells. Hand dug wells are the most inexpensive and innocuous provision of drinking water in As-contaminated rural areas using normalized present worth value during life cycle cost assessment (Chamberlain and Sabatini, 2014). They are an excellent choice to combat the water crisis in drying summer seasons in the shallow unconfined aquifer regions (Collins, 2000). Although, proper disinfection is mandatory to remove bacteriological contamination in the dug wells as the water quality varies seasonally and locally (Alam and Rahman, 2011). Chakraborti et al. (2015) reported that in As-endemic country like Bangladesh, dug wells could be used as alternate safe water source provided with proper construction. In the studied sites of Murshidabad, i.e. Raninagar-II block, dug wells at a depth of 9.14–21.4 m, seems to be uncontaminated as it showed a mean value of 3.78 ± 1.01 μg/L (range: <3–5.5 μg/L ; n = 5); no samples were found with As >10 μg/L . Mean Fe concentrations in dug well and pipeline supplied water samples were found to be 0.17 mg/L (range: <0.05–0.36 mg/L; n = 5)and 1.27 mg/L (range:<0.05–4.7 mg/L; n = 10), respectively which proved that the water quality of dug well is safer than pipeline supplied water with respect to Fe concentration **(Fig. 65)**.

Fig. 65. Dug wells being used in Domkol, Murshidabad

It is important to discourse here that Fe/As ratio is a significant factorfor As removal. Some amountof dissolved As gets removed by co-precipitation with the naturally occurring Fe (Mamtaz et al., 2001). But, higher Fe/As value is requiredto attain As-safe drinking water

(Banerji and Chaudhari, 2017; Schmidt et al., 2016). The studied dugwells has apparently high natural Fe/As ratio (mean: 66.1, range: 9.1–122). Mazumder et al. (2020) also found that the drinking water samples of dug wells in the studied As affected blocks of Maldah district were As safe. Chakraborti et al. (2015) also reported that in As-endemic country like Bangladesh, dug wells could be used as alternate safe water source provided with proper construction. On this note, the initiative taken by the neighboring state Bihar to restore 82,000 dug wells forprovidingsecurity about unpolluted drinking water is must appreciated which will ensure good health as well as lessen water scarcity during dry summer (The Telegraph, February, 22, https://www.telegraphindia.com/india/bihar-to-renovate-82000-dug-wells-to-provide-drinking-water-security/cid/1852882).

An elaborative water quality study has been done on selected 11 dug well samples to verify its potency for use in domestic and drinking purpose (**Table 60**). Water quality is neither acidic nor alkaline (average pH value is 7, range: 6.1 -7.5). Turbidity measures the way to the water loses its clearness owing to the existence of suspended materials. Turbidity of the dug well water samples vary between 1 NTU and 26 NTU (average: 5.5 NTU). The presence of sediment, suspended soil, carbon particle, inorganic substance, and other undetectableliving things enhances turbidity which can be removed by filtering through fine Maslin cloth at domestic level. The Electrical conductivity value is seemingly very high (average: 1183 µS/cm; range: 391-2250 µS/cm), i.e. the potential of water to carry minerals is very high. The TDS value ranges between 33 mg/L and 2700 mg/L with a mean value of 1121 mg/L. The regression value between TDS and EC (R^2= 0.77) signifies that there exists a very good relation between the dissolved solids and their conductance. The Cl^- content in dug well samples is moderate which suggests that the water is not salty; the average value (209 mg/L) is within the upper bound value of 250 mg/L (WHO, 2012). Usually, Cl^- is present in natural water in lower concentration. For the presence of Cl^- in dug well water, the accountable factors are the soil and rocks, atmospheric precipitation etc. Higher Cl^- concentration may be seen in summer due to reduction in water level (Patil et al., 2015). The SO_4^{2-} concentration range lies between 9.7 and 148 mg/L (average: 45.7 mg/L). The water quality is found to be very hard in nature with average of 455 mg/L (range: 160-940 mg/L). 90.9 % water sample is with higher water hardness than the recommendation by BIS. Hardness is caused by diverse dissolved polyvalent metallic ions, specifically calcium and magnesium cations. The hardness comes mainly from calcium content which is proved by the range of calcium concentration found in samples (20-272

mg/L). The average calcium concentration is found to be 100 mg/L, higher than the recommended value of BIS, 2012 (75 mg/L). 63.6 % water samples are found with higher calcium concentration than recommendation. Although, the permitted limit in absence of any alternative source is 200 mg/L. The Mg^{2+}concentration ranges between 4.9 and 117 mg/L with an average of 49.5 mg/L. The tolerable limit of Mg^{2+}concentration is 30 mg/L and permitted limit in absence of any alternate source is 100 mg/L. Calcium ion arise in water naturally through the dissolution of CO_3^{2-} minerals and the breakdown of the sulfate, phosphate, and silicate reserves while magnesium comes from dolomite and other magnesium containing compounds in sediments (Gebresilasie et al., 2021). The total alkalinity (as $CaCO_3$) value is observed as 74.5 mg/L (range: 30-100 mg/L), lower than recommendation (200 mg/L). Average carbonate and bicarbonate concentration of water samples are observed as 61.8 and 43.6 mg/L. Above all, it has been observed that As concentration range is <3 – 33.9 µg/L with an average concentration of 6.5 µg/L. Almost 81.8 % of studied samples are safe with respect to As concentration. Iron concentration is found within the range of <0.01 – 2.9 mg/L (average concentration: 0.2 mg/L). The Fe/As molar ratio range lies between 0.53 and 85.8 (average: 12.6).

Temporary hardness of water is mostly contributed by calcium bicarbonate which can be removed by boiling. Boiling helps in precipitation of the dissolved minerals from the water. In view of the fact that boiling removes the calcium content of water, it produces softer water. It is a rapid and inexpensive method to make hard water potable. However, boiling works only on temporary hardness, not permanent hardness asthe secondone contains dissolved calcium sulfate that boiling does not remove. As the sulphate concentration is observed to be low, the water hardness can be termed as temporary hardness. Permanent hardness of water is removed by lime softening using hydrated calcium oxide or lime soda (lime plus sodium carbonate). Disinfection of water can be performed using $KMnO_4$ or zeolite. Generally, $KMnO_4$ controls the odor and taste of water while zeolite controls bacteria, viruses, fungi and algae (Hira Smith et al., 2003). Therefore, with proper maintenance and minimum treatment at domestic level, the dug well water can be consumed easily.

Table 60: Physico-chemical parameters of the dug well water samples of Domkal, Murshidabad

Dug well Sl no.	Village	Block	Depth (ft)	pH	Turbidity (NTU)	EC (µS/cm)	TDS (mg/L)	As (µg/L)	F^- (mg/L)	Fe (mg/L)	Cl^- (mg/L)	SO_4^{2-} (mg/L)	NO_3^- (mg/L)	Total hardness (mg/L)	Ca (mg/L)	Mg (mg/L)	Total Alkalinity (mg/L)	CO_3^{2-} (mg/L)	HCO_3^- (mg/L)
1	Damodarpur	Raninagar-I	50	7.35	1	1841	2700	<3	0.092	0.025	515	148		940	272	63.3	80	60	50
2	Pomaipur	Raninagar-I	45	7.32	2	932	567	<3	0.23	<0.01	245	49		410	88	46.2	70	100	20
3	Madanpore	Raninagar-I	45	7.31	2	391	33	<3	0.057	<0.01	96	14.9		160	28	21.9	30	60	0
4	Nabipore	Raninagar-II	50	7.52	5	2250	1833	<3	0.22	<0.01	440	77.7		730	100	117	110	60	80
5	Natiyal	Raninagar-II	40	6.89	7	1054	800	33.9	0.3	2.91	107	15.6		360	20	75.4	100	0	100
6	Radhakantapur	Domkol	37	7.32	3	634	600	18.7	0.33	<0.01	131	52.7		280	80	19.5	50	60	20
7	Radhakantapur	Domkol	40	6.05	26	1122	1100	3.24	0.43	<0.01	178	12.8		570	140	53.5	100	80	60
8	Battanabad	Domkol	45	7.11	6	1125	1033	<3	0.14	<0.01	106	9.65		380	92	36.5	80	40	60
9	Kalukup	Domkol	40	6.64	3	1478	1366	<3	0.064	<0.01	249	56.6		450	172	4.9	60	100	10
10	Kalukup	Domkol	42	6.9	3	1208	1166	<3	0.056	<0.01	142	33.4		310	60	38.9	50	40	30
11	Kalukup	Domkol	40	6.83	2	984	1133	<3	0.052	<0.01	96	32.1		420	56	68.1	90	80	50
	Avg.		43.1	7.0	5.5	1183	1121	6.5	0.2	0.2	209	45.7		455	100	49.5	74.5	61.8	43.6
	Min		37.0	6.1	1.0	391	33.0	<3	0.1	<0.01	96.0	9.7		160	20.0	4.9	30.0	-	-
	Max		50.0	7.5	26.0	2250	2700	33.9	0.4	2.9	515	148		940	272	117	110	100	100

5.6.4. Risk assessment among all the available five different sources of drinking water in Raninagar-II, Murshidabad

Considering all the available sources of drinking water in the studyarea, another health risk assessment study of As toxicity has been donefollowing eq. (13) based on a severity adjustedmargin of exposure (SAMOE) approach.

Table 61: Risk thermometer of all kind of available drinking water sources

Drinking water source	Mean As concentration (µg/L)	Daily intake of As (µg/day) (Adult = 5 l/day)	Daily Exposure or 'E' (µg/kg bw/day) [BW_{Adult} = 60 kg]	SAMOE value	Risk Class
Domestic and community level shallow tube-well	63.8	319	5.32	0.006	Class 5 (high)
Pipeline supplied water	21	105	1.75	0.017	Class 4 (moderateto high risk)
Sajaldhara treatment plant	13.6	68	1.13	0.026	Class 4 (moderateto high risk)
Reverse Osmosis plant	7.76	38.8	0.65	0.046	Class 4 (moderateto high risk)
Dug well	3.78	18.9	0.32	0.1	Class 3 (low to moderate)

The estimated SAMOE values and risk level of As toxicity among all kind of drinking water sources have been shown. The evaluation shows that drinking water from domestic and community tube-wells accounts highest health risk for adults (class 5, SAMOE: 0.006) while dug well is apparently the safest source of drinking water as the estimated SAMOE value is 0.109 (Class 3; risk: 'low to moderate'). Rest of the three available sources of drinking water in the studied areas shows 'moderate to high risk' as they fall in 'Class 4'. The reference risk categories described by Sand et al., (2015a, b) are also given in **Fig. 66** to compare with our findings.

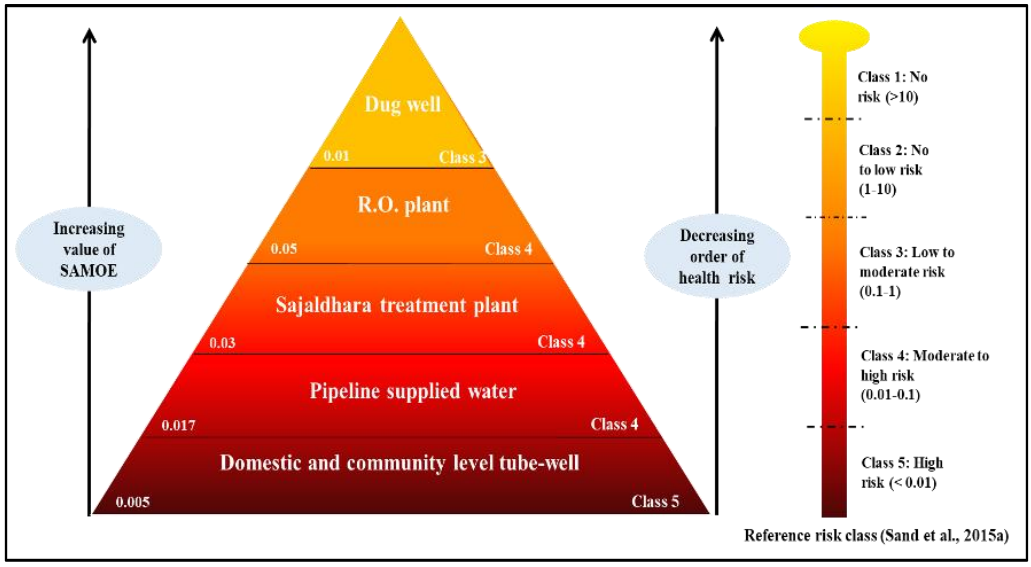


Fig. 66. Assessment of health risk from all kind of available drinking water sources

5.6.5. Application of solar oxidation in removal of arsenic (SORAS) from contaminated drinking water using iron and Vit-C enriched sources

There is no such successful and easy to operate As removal process opted in household level. In most of the useful Asremoval technologies oxidation is one of the primary steps. Arsenite is generally present in groundwater as neutral molecules below pH 9.2, whereas, arsenate occurs as anions in the pH range 4–10 (Masscheleyn et al., 1991). Most treatment methods like coagulation, adsorption or ion exchange are effective in removing arsenate and hence include 'oxidation' of arsenite to arsenate as a pre-treatment step (Ghurye and Clifford 2004; Leupin and Hug 2005). To remove As from the solution oxidation must be coupled with a removal process likeadsorption, coagulation or ion exchange. Oxidation mainly refers to chemical oxidation that includesnumeral oxidizing agents such as permanganate, manganese dioxide, ozone, free Cl_2, hypochlorite, H_2O_2, Fenton's reagent (H_2O_2 /Fe^{2+}) and UV radiaton etc. (Jekel 1994; Kim and Nriagu 2000).

$$H_3AsO_3 + ½\ O_2 = H_2AsO_4^- + 2\ H^+$$

$$H_3AsO_3 + HClO = HAsO_4^{2-} + Cl^- + 3H^+$$

$$3H_3AsO_3 + 2KMnO_4 = 3HAsO_4^{2-} + 2MnO_2^{+} + 2K^+ + 4H^+ + H_2O$$

In-situ oxidation of As and Fe in the underground water has been trialed under the Danida Arsenic Mitigation Pilot Project in 2001. It involves storing the oxygenated tube-well water in a reservoir and releasing it back into the aquifers by opening a valve in a pipe that connects the water tank with the tube-well pipe under the pump head. The dissolved oxygen in water oxidizes As-III to less mobile As-V and also the Fe^{2+} to Fe^{3+}, ensuing a

decreased As concentration in tube-well water. The probable reactions of arsenate to ferric hydroxide [$Fe(OH)_3$] are given below.

$Fe(OH)_3$ (s) + H_3AsO_4 = $FeAsO_4.2H_2O$ + H_2O

$FeOH^o + AsO_4^{3-} + 3\ H^+ = {}^oFeH_2AsO_4 + H_2O$

Passive Sedimentation refers to aeration of water during collection and storage in containers that might initiate a reduction in Asconcentration in water which is called 'Bashi Pani'. The technique gained much attention due to the habit of rural people for drinking stored water from pitchers. Researches from Bangladesh showed zero to high reduction in As concentration by this process; although, reduction of As is seen through co-precipitation in presence of natural iron in specific hydro-geological condition. Most of the studies observed reduction of 0 - 25% of the initial As concentration in groundwater. However, passive sedimentation was unsuccessful in reducing Asupto the desired level of in any tube-well (Ahmed et al., 2001).

Co-precipitation is applied most regularly to remove As from contaminated water in several pilot-scale projects. The general coagulants used are alum ($Al_2(SO_4)_3.18H_2O$), ferric sulfate ($Fe_2(SO4)_3.7H_2O$) and ferric chloride ($FeCl_3$), active in removal of As from water (Edwards, 1994; Hering et al., 1997). Alum is most successful for the removal of As, if an oxidizing agent, like$Cl_2$is added before the flocculator and clarifier and the pH is decreased to 7 or less. While using iron salts, the As combines with the iron to form a precipitate (FeOOH) that settles out in the clarifier (sludge) followed by a filter. $FeCl_3$ is more commonly used instead of $FeSO_4$ and Fe^{2+}delivers an effective treatment giving residual dissolved As concentrations below1mg/L over a range of compositions with 99.9% removal and Fe^{3+}treatment is more effective in combination with mixed lime and $Mg(OH)_2$. The As removal through lime softening is significantly ruled by the pH and Cl_2. Chlorine is necessary to oxidize theAsand acid would possibly be required to reduce the pH of the treated water to make it potable (Choong et al., 2007).

The process of "Solar oxidation and removal of As (SORAS)" is actually a photochemical oxidation of As (III) followed by precipitation or filtration of As(V) adsorbed on Fe(III)oxides. They concluded that removal efficacy is regulated by the variation in the chemical matrix, or in the operational conditions. SORAS is a simple, economical household-level method for removing As from contaminated water in a moderate way (Hug et al. 2001; Hug and Canonica et al., 2001). UV light promoted As(III) oxidation when $H_2O_2$is present (Yang et al., 1999). In presence of sunlight Fe-II is oxidized to Fe-

III and in presence of some organic acid, the Fe-III (acidic complex) produces some radical (H_2O_2) or hydroxide radical or oxygen radical etc. which precipitates As with iron.

$H_2O_2 + h\gamma \rightarrow OH^{\bullet}$

$Fe(II) + O_2 \rightarrow Fe(III)$

Fe(III) + organic acid → Fe(III)–organic complex

Fe(III)–organic complex (irradiation with UV-A light) → organic radical + CO_2 + Fe(II)

Organic radical + $O_2 \rightarrow O_2^{\bullet -}$ + oxidized organic species

$O_2^{\bullet -} + Fe(II) \rightarrow Fe(III) + H_2O_2$

Throughout the world, several researches took place for As removal using sunlight. Arsenic oxidation and simultaneous removal from water were evaluated with use of TiO_2 immobilized in PET containers in presence of sunlight and iron salts (Fostier et al., 2008). After treating with 10 % TiO_2 coating solution, Fe(II): 7.0 mg/L and 2 h of solar irradiation, 99% removal was observed. Garcia et al. (2004) suggested that, to de-arsenificate contaminated water of unknown composition, the best practice is the use of moderate amounts of iron andlittle concentrations of citric acid at direct sunlight. They explained that oxides generated by ferrous salts are more competent in comparison to the solids formed by Fe(III) hydrolysis. Moreover, bicarbonate alkalinity is also essential to allow the sufficient precipitation. Adding little doses of citric acid source is advantageous, but with higher concentrations, the result may not come, due to possible interference of solid formation. In another experiment, As(III) seems to be oxidized photo-chemically within 2-3 hrs by irradiation with 90 W/m^2 UV-A light.Fe(III)-citrate complexes catalyzed oxidation of As(III). 3-oxoglutaric acid, the photo-product of citrate instigatedfast flocculation and precipitation of Fe(III). During experiment in laboratory, after addition of 50 μM citrate or 100-200 μL oflemon juice/L, 80-90% of totalAswas removed with radiation for 2-3 h and precipitation. Thesame practice was capable to take out 45-78% of As in raw groundwater (Hug et al., 2001). Researchers say that In Camarones, Chile, As removal efficiency through SORAS has been observed 99 % in synthetic as well as in natural waters whereas In Tucumán, Argentina As removal efficiency has been observed by 90% in synthetic waters but maximum 60% is observed in natural groundwater (dHiriart, et al., 2009). Application of SORAS for As safe drinking water in several countries throughout the world have been defined in **Table 62**.

Table 62: Application of SORAS in different countries and their results

Country	Initial Fe concentration	Initial pH of water	Initial As concentration	Final As concentration	Final pH of water	Additional element added	Arsenic removal %	Reference
Bangladesh	5 mg/L (Fe-II)	7.0-7.2	500 µg/L (As-III)	<50	7.0	Lemon juice (100-200 µl)	50-70	Wegelin et al., (2000)
Chile	<0.10 mg/L	8.3	1040 µg/L	4.8	7.8	Steel wool (1.3 g/l) & lemon juice (4.5 mg citrate/l)	>99.5	Cornejo et al., (2008)
Argentina	3 mg/L	8.56	1023 µg/L	753	7.57	Lemon juice 750 µl/l	26	Garcia et al., (2004)
		8.73	614 µg/L	372	7.61		39	
		8.39	210 µg/L	147	6.27		38	
		7.7	88 µg/L	37	7.39		58	
Ireland	12 mg/L	-	1000 µg/L	<10	-	Citrate addition showed detrimental effect	20-90	O'Farrell et al., (2016)
Bangladesh	0.06-5 mg/L	6.5-8.0	500 µg/L	-	6.5-8.0	50 µM citrate or 100-200 µl	80-90	Hug et al., (2001)
India	5 mg/L	7.2-7.7	250 µg/L	<50	7.4-8..3	50 µM citrate & tartarate	80	Majumder and Chaudhuri, (2005)
Chile	Fe-II: 125.3 µmol/l	6.80	6.7 µmol/l	-	-	Citrate 30 µmol/l	>80	Lara et al., (2006)
	Fe-II: 89.5 µmol/l	6.80	6.67 µmol/l	-	-	Citrate 50 µmol/l	>80	
India	2-6 mg/L	-	100-500 µg/L	10-110	-	Tomato juice 0.5 -1 ml/l	78-98.2	Majumder et al., (2013)
	2-6 mg/L	-	100-500 µg/L	50-310	-	Lime juice 0.5 -1 ml/l	39-69.2	

5.6.5.1. Iron, Vit-C, Proline and micro-nutrient content of the targeted components

Few common fruits and seeds have been chosen with respect to their nutritional and anti-toxic properties. Amla, Pomegranate, Apple, Guava, Orange, and Tomato were targeted as a natural iron and vitamin-C enriched fruit sample. Ascorbic acid (vitamin C source) found in food naturally. The ascorbic acid present in these fruits boost the Fe content in the body systems controlling the blood count. Amla (Indian gooseberry) is extensively

grown in tropical and sub-tropical regions and has therapeutic prospective against lethal diseases. It is enriched with vitamin-C, polyphenols like tannins, ellagic acid, gallic acid, calcium, iron, flavonoids like quercetin and rutin, essential amino acids and several other minerals and anti-oxidants (Kulkarni and Ghurghure, 2018). Pomegranate is a strong antioxidant, rich in flavonoids, punicic acid, alkaloids, anthocyanins, mono and di-saccharides, simple organic acids and it also has anti-inflammatory and antihypertensive properties (Lansky et al., 2000). Apples contain antioxidants that can hinder the growth of cancer cells; shield the cells in pancreas lowering the probability of type 2 diabetes (Hyson, 2011). Tomato and orange, as citrous fruits, are enriched with b-carotene and ascorbic acid, potassium, calcium etc. (Paul and Shaha, 2004). Guava is high in antioxidants like phenols and ascorbic acid (Lim et al., 2006). Bean, Pumpkin seed, Sesame seed, and lentils (Mung daal and Massor daal) were targeted as a natural iron and vitamin-C enriched grain samples. Pumpkin seeds contain substantial amounts of different types of amino acids but low amounts of calcium, iron and other essential trace elements (Glew et al., 2006). Lentils are enriched with protein, fiber, vitamin B1, minerals, and essential amino acids and an excellent source of selenium depending upon cultivation land (Sah and Smits, 2013). Proline is a vital osmolyte that reduces the membrane injuredone by oxidative stress; takes care about proteins against denaturation and water balance under As stress, reported by Schat et al. (1997). Therefore, estimated iron, Vit-C and proline (amino acid) content of the targeted components are given in **Table 63**.

Table 63: Iron, Vit-C, Proline content of all the targeted components

Sl.no	Component name	Iron (mg/100 g)	Vit-C (mg/100 g)	Proline (mg/g FW)
1	Moong dal (lentils)	6.1	4.8	4.18
2	Masoor dal (lentils)	3.1	1.5	7.74
3*	Pumpkin seed	3.5	0.4	18.4
4*	Sesame seed	14.6	-	1.50
5	Beans	1.4	5.8	3.21
6	Tomato	0.77	23.3	1.46
7	Guava	0.3	333	3.48
8	Apple	0.1	4.5	1.55
9	Orange	0.32	66.6	4.05
10*	Amla	1.97	516	5.77
11*	Pomegranate	1.38	60.4	2.52

*selected components

It is observed that the iron content was found to be maximum in sesame seeds (14.6 mg/100 g) followed by lentils, pumpkin seed (3.5 mg/100 g), amla (1.97 mg/100 g) and pomegranate (1.38 mg/100 g). Vit-C concentration was found to be highest in amla (516 mg/100 g) followed by guava (333 mg/100 g), orange, pomegranate etc. The grains have lower content of ascorbic acid compared to the citrus fruits. The order of proline content was like: pumpkin seeds> lentils > amla > orange followed by guava. The lowest content of proline was observed in sesame seeds (1.50 mg/g). On a cumulative approach, amla, pomegranate, pumpkin seed, and sesame seed) were chosen as potential ingredients for possible de-arsenification from drinking water applying solar oxidation method. In a previous study, the single polynomial function revealed that theiron concentration was the most significant factorin the removal of As by precipitation. Theoptimal molar ratio for As, citrate and Fe was 1:4.5:18.7, when more than 90% ofAs being eradicated after 4 h of irradiation period (Lara et al., 2006).

It is well established fact that selenium (Se) and zinc (Zn) are anti toxic to As. Zn is known for increasing the level of As detoxifying protein metallothionein (Roychowdhury et al., 2003). In Asexposed areas, the affected people are conventionally administered with nutritious food containing vitamin C, α-tocopherol, flavonoids, polyphenols and anti-toxic micronutrients like Se and Zn (Rahman et al., 2019; Spallholz et al., 2004). So, Se and Zn concentrations have been estimated for the selected materials (**Table 64**). Surprisingly, it has been observed that, both the seeds have higher amount of micronutrients concentration than the fruit juices which are more enriched with ascorbic acid. Sesame seeds have highest selenium and zinc concentration among the selected components (32.0 and 7.19 µg/100 g) respectively. Therefore, it is going to be a beautiful study where the potential of micronutrients, ascorbic acid and iron concentration will be compared and evaluated in de-arsenification process.

Table 64: Micronutrients concentration (Selenium, Zinc) in the selected components

	Amla (µg/100 g)	**Pomegranate (µg/100 g)**	**Pumpkin seeds (µg/100 g)**	**Sesame seeds (µg/100 g)**
Selenium	0.65	0.42	9.0	32.0
Zinc	0.12	0.35	7.03	7.19

5.6.5.2. Solar Oxidation with different doses of components on varying As-concentrated spiked water

Synthetic solutions were prepared in the laboratory by spiking required As concentrations (500 µg/L, 250 µg/L, 100 µg/L) from 1000 µg/L stock solution of As-III and As-V in 500

ml transparent PET bottles and made the volume up with tap water. Each solution were filled in eight separate PET bottles. The solutions were subjected to different dose of amla and pomegranate (10 ml/l and 20 ml/l) and different doses of pumpkin seeds and sesame seeds (2 g/L and 4 g/L) and mixed for 15 minutes with an orbital shaker. This mixing provides sufficient oxygen for oxidation of Fe(II) to Fe(III). For preparation of synthetic solutions, tap water was used whose iron concentrations were estimated to be 0.25 mg/L. The PET bottles were kept in direct sunlight for 4 long hours (12 pm to 4 pm). The experiment was done during winter season (January to February, 2020). Experimental samples were collected twice from the distinct containers through decanting the bottles after 2hr and 4 hr respectively. Changes in As and Fe concentration was estimated with different time irradiation. Samples were collected twice after 2 hr and 4 hr and preserved with nitric acid and stored in the fridge at 4 °C. Arsenic and Fe concentration were measured and described in the following table (**Table 65**). pH of the water samples at two times has been evaluated also. A blank experiment has been performed without addition of any external component; only passive sedimentation has been checked.

Table 65: As-spike water subjected to different dose of added components

			2hr		4 hr	
Initial As concentration (µg/L)	**Component**	**Dose (ml/l or g/l)**	**As concentration (mg/L)**	**Fe concentration (mg/L)**	**As concentration (mg/L)**	**Fe concentration (mg/L)**
500	Amla	10	343	2.25	300	1.46
		20	321	2.75	280	0.98
	Pomegranate	10	458	2.07	345	0.99
		20	446	2.3	320	1.03
	Pumpkin seeds	2	432	1.13	390	0.77
		4	407	2.2	356	1.62
	Sesame seeds	2	448	1.01	410	0.7
		4	434	1.1	401	0.79
250	Amla	10	158	2.49	110	1.04
		20	182	2.35	114	1.08
	Pomegranate	10	220	2.44	158	0.77
		20	210	2.22	127	0.74
	Pumpkin seeds	2	202	0.83	175	0.58
		4	222	1.03	198	0.88
	Sesame seeds	2	209	1.33	200	0.76
		4	195	1.53	188	0.86
100	Amla	10	72	1.98	48	0.99
		20	87	2.15	55	0.81
	Pomegranate	10	88	2.07	52	0.65
		20	82	2.13	67	0.73
	Pumpkin seeds	2	86	0.78	75	0.45
		4	97	0.84	77	0.54
	Sesame seeds	2	91	1.05	88	0.31
		4	94	0.87	85	0.32

5.6.5.3. Arsenic removal percentage

The removal percentage of As was calculated according to equation:

$$\text{Arsenic removal (\%)} = (C_i - C_f)/ C_i \times 100 \qquad \text{eq. (19)}$$

Where, C_i is the initial As concentration (mg/L) and C_f is the final As concentration (mg/L) after 2 and 4 hour.

5.6.5.3.1. Arsenic removal percentage with varying arsenic concentration of initial solution

The variation in As concentrations during different time of irradiation at three types of initial spiked solutions is shown in **Fig.67a, b and c.** Also, the average As reduction percentage values at three types of initial spiked solutions are shown in **Fig. 67d**. It appears that the As reduction percentage order follows like amla > pomegranate > pumpkin seeds > sesame seeds at all the three solutions. Fig shows that after 4h, As reduction is best observed with 250 and 100 μg/LAs concentrations (Fig. 67c & 67b). It is observed that the capacity of As reduction of amla is highest for all the experimental conditions. After final irradiation period, at the three different types of initial As concentrated solutions i.e. 500, 250 and 100 μg/L, the observed average As reduction percentage of amla was 42, 55.2 and 48.5 % including two different doses **(**Fig. 67d**)**. At the same condition, the observed average As reduction percentage of pomegranate was found to be 33.5, 43 and 40.5 %. It indicates that the reduction percentage for both the component was greatest at 250 μg/L As concentration. For amla, at dose-I (10 ml/L), the reduction percentages are 40, 56 and 52 at 500, 250 and 100 μg/L solutions while at dose-II (20 ml/L), the reduction percentages are 44, 54.4 and 45 %. Similarly, for pomegranate, at dose-I (10 ml/L), the reduction percentages are 31, 36.8 and 48 at 500, 250 and 100 μg/L solutions while at dose-II (20 ml/L), the reduction percentages are 36, 49.2 and 33 %. Likewise, after final irradiation period, at the three different type of initial As concentrated solutions i.e. 500, 250 and 100 μg/L , the observed average As reduction percentage of pumpkin seeds were stagnant i.e. 25.4, 25.4 and 24 % including two different doses. The observed average As reduction percentages of sesame seeds at the same experimental conditions were found to be the lowest of all; 18.9, 22.4 and 13.5 %. It indicates that the reduction percentage for pumpkin seeds were almost same at three Asconcentrations whereas the reduction percentage for sesame seeds were maximum at 250 μg/LAs concentration. For pumpkin seeds, at dose-I

(2 g/L), the reduction percentages are 22, 30 and 25 % at 500, 250 and 100 µg/L solutions while at dose-II (4 g/L), the reduction percentages are 28.8, 20.8 and 23 %. Similarly, for sesame seeds, at dose-I (2 g/L), the reduction percentages are 18, 20 and 12 % at 500, 250 and 100 µg/L solutions while at dose-II (4 g/L), the reduction percentages are 19.8, 24.8 and 15 %. Therefore, it can be said that in this process of solar oxidation and remediation, As reduction is not sufficient at higher initial As concentrations and the fruit juice samples are more competent than the seed dust samples. However, Majumder and Chaudhuri (2005) reported that with increase in initial As concentration in solution up to 1000 µg/L , removal efficiency decreased when Fe concentration was 5 mg/L. they used citrate-tartarate combined which was able to remove As below 50 µg/L under 6-8 h of irradiation period.

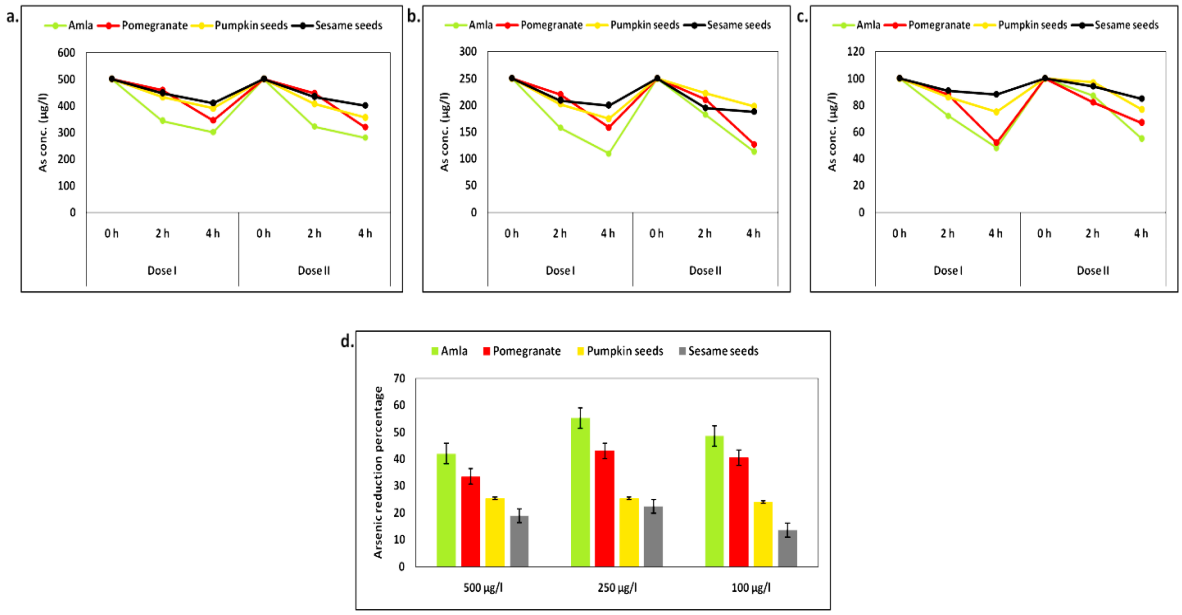


Fig. 67. Variation of arsenic concentrations at three types of initial concentrated spiked solutionsduring different time of irradiation

5.6.5.3.2. Arsenic removal percentage with varying irradiation time

De-arsenification capacity increases with increasing irradiation time with sunlight. On an average of the three different types of solutions, the As reduction percentage is highest for amla at two doses. At dose I, the observed reduction percentages for amla, pomegranate, pumpkin seeds and sesame seeds at 4 h of irradiation period are 49.3, 38.6, 25.7 and 16.7 % (**Fig. 68a**). The reduction percentages have been increased from 2h by almost 1.54, 3.57, 1.65 and 1.40 times respectively. At dose II, the observed reduction percentages for the

four components at 4 h of irradiation period are 47.8, 39.4, 24.2 and 19.9% respectively (**Fig. 68b**). The reduction percentages have been increased from 2h by approximately 1.89, 2.64, 2.21 and 1.45 times. Considering both the doses, the average Asreduction percentages for the four components were observed as 48.6, 39.0, 24.9 and 18.3 % respectively which are higher by 1.69, 3.03, 1.88 and 1.42 times from 2h. SORAS remove As in a two-step procedure. Firstly, As(III), which only weakly adsorbs to iron(hydr)oxides, is oxidized to the strongly adsorbing As(V). Next, Fe(III)(hydr)oxides formed from naturally present iron are settled at the bottom of the container with the adsorbed As(V) and the clear water is decanted and filtered with a fine cloth. Instead of adding chemical oxidants such as chlorine or permanganate, reactive oxidants are produced photo-chemically with sunlight.

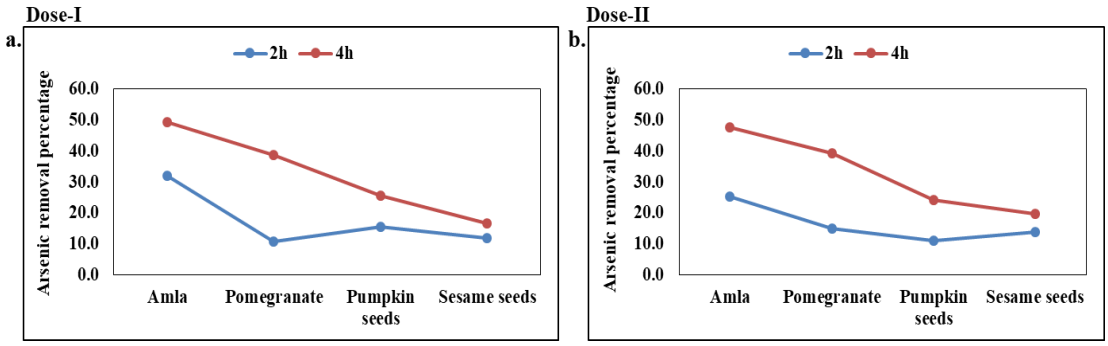


Fig. 68. Arsenic removal percentage with at two different doses with reference to irradiation time

5.6.5.3.3. Arsenic removal percentage with varying dose

It is observed that addition of vit-C enriched components lead to a much faster formation of precipitates and settling than the blank experiment. When dose is constant, with increasing irradiation time, As removal percentage increases. At dose-I (fruit juice: 10 ml/L, seed dust: 2 g/L), after 2h of irradiation, As removal efficiency is maximum for amla and lowest for pomegranate at 500 and 250 µg/L initial Asconcentrated solutions, while at 100 µg/L solution; the efficiency remains same for amla but seems to be lowest for sesame seeds. At 4h of irradiation time, As reduction percentage is highest for amla followed by pomegranate, pumpkin seeds and sesame seeds. At 500 µg/L initial As concentrated solution, for amla and pomegranate, As reduction percentage increases by 1.27 and 3.69 times respectively with increasing time of radiation. Similarly, the percentage increase for amla and pomegranate at 250 and 100 µg/Linitial As concentrated solutions are 1.52, 3.06

and 1.85, 4 times respectively **(Fig. 69).** At seed dust dose of 2 g/l, the percentage reduction also increases with time. At varying initial As concentrated solutions (500, 250 and 100 μg/L), As removal percentage increases by 1.62, 1.56, 1.78 times for pumpkin seeds and 1.73, 1.22, 1.33 times for sesame seeds respectively. At 10 ml/l fruit juice dose and at 4 h of irradiation, the average As removal efficiency is observed to be higher for amla (49.3%, range: 40-56 %) than pomegranate (38.6 %, range: 31-48 %). At 2 g/L seed dust dose and at 4 h of irradiation, the average As removal efficiency is observed to be higher for pumpkin seeds (25.6 %, range: 22-30 %) followed by sesame seeds (16.7%, range: 12-20 %).

At dose-II (fruit juice: 20 ml/L, seed dust: 4 g/L), after 2h of irradiation, As removal efficiency is maximum for amla at 500 and 250 μg/L initial Asconcentrated solutions (35.8 and 27.2 % respectively). However, at 100 μg/L solution, the efficiency is highest for pomegranate (18 %). Arsenic reduction potential appears to be lowest for pumpkin seeds (3 % and 11.2 %) at 100 and 250 μg/LAs concentration but it ranks 2nd highest at 500 μg/L solution. At 4h of irradiation time, As reduction percentage is highest for amla followed by pomegranate, pumpkin seeds and sesame seeds when initial As concentration is 500 and 100 μg/L. When initial As concentration is 250 μg/L, As reduction percentage after 4 h of irradiation is highest for amla followed by pomegranate, sesame seeds and pumpkin seeds. At 500 μg/L initial As concentrated solution, for amla and pomegranate, As reduction percentage increases by almost 1.23 and 3.33 times respectively with increasing time of radiation. Similarly, the percentage increase for amla and pomegranate at 250 and 100 μg/L initial As concentrated solutions are nearly 2, 3.07 and 3.46, 1.83 times respectively. At seed dust dose of 4 g/l, the percentage reduction also increases with time. At varying initial As concentrated solutions (500, 250 and 100 μg/L), As removal percentage increases by 1.55, 1.86, 7.67 times for pumpkin seeds and 1.5, 1.13, 2.5 times for sesame seeds respectively. At 20 ml/l fruit juice dose and at 4 h of irradiation, the average As removal efficiency is observed to be higher for amla (47.8%, range: 44-54.4%) than pomegranate (39.4 %, range: 33-49.2%). At 4 g/l seed dust dose and at 4 h of irradiation, the average As removal efficiency is observed to be higher for pumpkin seeds (24.2%, range: 20.8-28.8%) followed by sesame seeds (19.9 %, range: 15-24.8%). Finally, it can be said that dose-I is more effective than dose-II for amla and pumpkin seeds with reduction percentage of 49.3 and 25.6%, while dose-II is more effective for pomegranate and sesame seeds with reduction percentage of 39.4 and 19.9%. With an average of two different doses, it can be said that As reduction efficiency is highest for amla (48.5%),

followed by pomegranate, pumpkin seeds and sesame seeds (39, 24.9 and 18.3%) respectively. Although the reduction potential is apparently low, but it can be increased when the water iron concentration will be high.

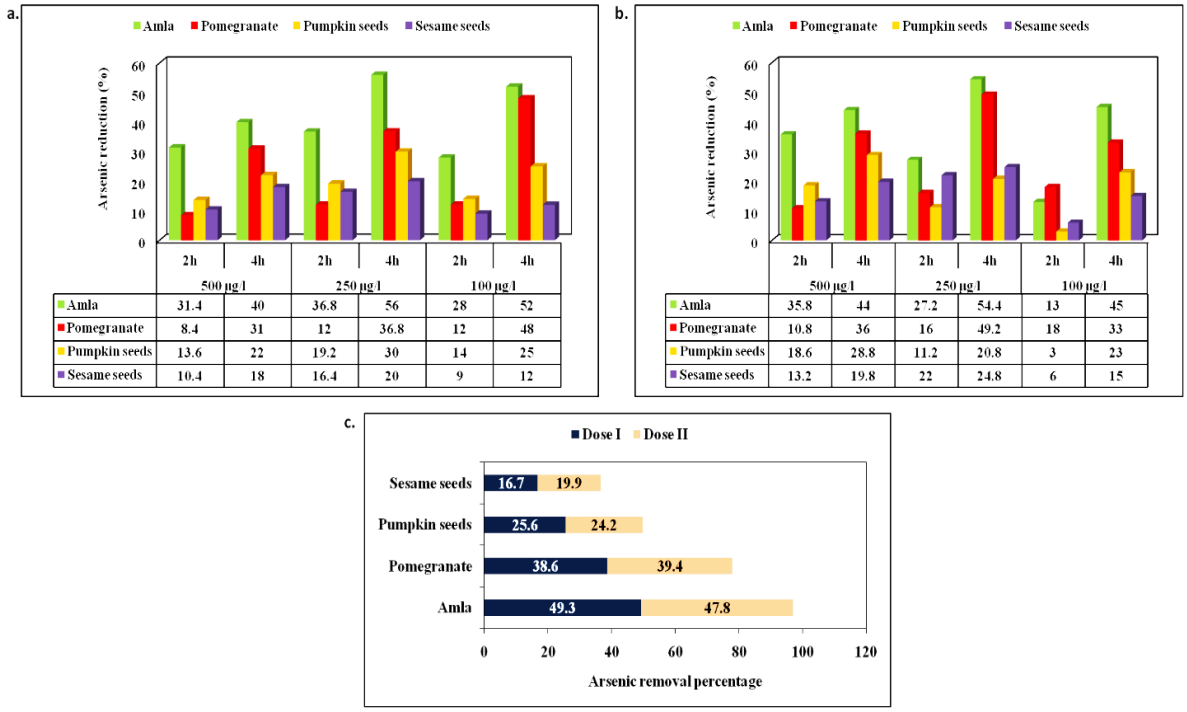


Fig. 69. Arsenic removal percentage with reference to difference doses

In another study in Chakdah block of Nadia district, West Bengal, groundwater samples from domestic hand tube wells were taken for SORAS treatment with application of synthetic Fe salt or tablets followed by addition of different citrate doses. Lime, tomato and lemon were used as citrate source and the bottles were exposed to sunlight for 4 h. Arsenic removal efficiencies of tomato, lemon and lime were reported to be 88%, 73% and 50%, respectively. The Arsenic removal efficiency of tomato was found to be higher than lime and lemon (Majumder et al., 2013). Few other experiments have been performed in rural Bengal in SORAS using lemon juice as citric source. It revealed that approx. 3 - 10 drops of lemon juice per litre water should be added instantly after filling the bottle with pumped groundwater. A too high concentration of lemon juice reduces the efficiency of solar oxidation process as well as taste of water. They claimed that SORAS-treated water is clear and doesn't change the taste. Food cooked with this treated water keeps its natural colour and freshness, e.g. cooked rice and vegetables are no longer of a red-brown colour. People living in As-affected areas seem to be eager to use the SORAS treatment method. However, lemons are not always available and the cleaning of bottles is somewhat burdensome. They suggested that $KMnO_4$ could be used during low sunlight radiation or

non-availability of lemons and coating of the inner walls of the bottles by iron particles which can be avoided by not completely filling the plastic bottle (Wegelin et al., 2000). In this perspective, the present study is important so that these organic materials can be used in exchange of lemon juice. Cornejo et al. (2008) showed high As removal efficiency in natural waters with use of zero-valent iron combined with citrate dose and solar radiation. The researchers used commercially available steel wool for additional iron source and lemon juice as citrate source. The maximum As removal was observed at 6 h of irradiation by using 1.3 g of zero-valent iron and 4.5 mg of citrate per liter. Arsenic removal percentages were observed higher than 99.5% and the final As concentration is below 10 mg/L. It clearly proves that to reach above 90% efficiency, iron concentration should be very high in the water.

5.6.5.4. Iron concentration in treated water

The tap water iron concentration is estimated to be 0.09 mg/L. So, the final iron concentration in the decanted water at different time of illumination with sunlight is the summation of tap water iron concentration and leached iron from the added materials. The decanted water had higher iron concentration at 2h of irradiation than 4h which signified the possibility of precipitation of iron with As.It is seen that after total irradiation period, iron concentration decreases in the solutions. The average iron precipitation values (dose I and II) are highest for pomegranate in the three solutions and lowest for pumpkin seeds. The iron precipitation percentage average values for amla are 49.7 (35.1-64.4%), 56.1 (58.2-54.0%) and 56.2 (50-62.3%) at 500 µg/L, 250 µg/L, and 100 µg/L respectively. In other words, the iron co-precipitation increases by 45.4 and 19.8 % in 500 and 100 µg/L solutions while it decreases by 7.8 % in 250 µg/L solution (**Fig. 70**). The iron co-precipitation percentage average values for pomegranate are 53.7 (52.2-55.2%), 67.6 (68.4 – 66.7%) and 67.2 (68.6-65.7%) at 500 µg/L, 250 µg/L, and 100 µg/L respectively. The iron co-precipitation percentage average values for pumpkin seeds are 29.1 (31.9-26.4%), 22.3 (30.1 – 14.6 %) and 39 (42.3-35.7%) at 500 µg/L, 250 µg/L, and 100 µg/L respectively. For sesame seeds, at the two doses, the iron precipitates by 30.7 and 28.2% (average: 29.4%) in 500 µg/L, by 42.9 and 43.8 % (average: 43.3%) in 250 µg/L and by 70.5 and 63.2% (average: 66.8%) in 100 µg/L solutions. It can be said that the precipitation percentage of iron capacity is larger when As concentration is comparatively low which denotes, to enhance As reduction, iron concentration needs to be high in solutions.

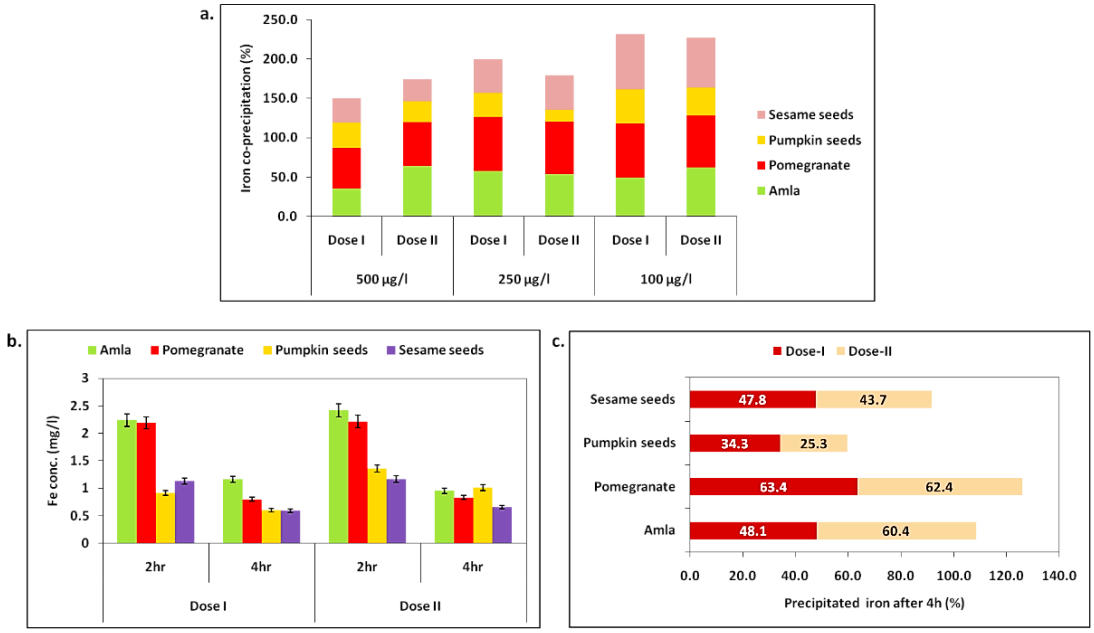


Fig. 70. Iron concentration of the treated water at different experimental variation

It is observed that the average iron concentrations in water solutions for three types of spiked solutions are decreased from 2h to 4h at both the doses. At dose I, initially after 2h, the average iron concentration for amla, pomegranate, pumpkin and sesame seeds are 2.24, 2.19, 0.91 and 1.13 mg/L respectively. With time, it decreased into 1.16, 0.80, 0.6 and 0.59 mg/L respectively. The decreasing trend signifies that the iron gets precipitated with As. Therefore, it can be said that at dose-I, the iron co-precipitation percentages are 48.1, 63.4, 34.3 and 47.8 respectively (**Fig. 71**). Similarly, at dose II, after 2h, the average iron concentration for amla, pomegranate, pumpkin and sesame seeds are 2.42, 2.22, 1.36 and 1.17 mg/L respectively. With time, it decreased into 0.96, 0.83, 1.01 and 0.66 mg/L respectively. The decreasing trend signifies that at dose-II, the iron co-precipitates at 60.4, 62.4, 25.3 and 43.7%, respectively (Fig. 71).

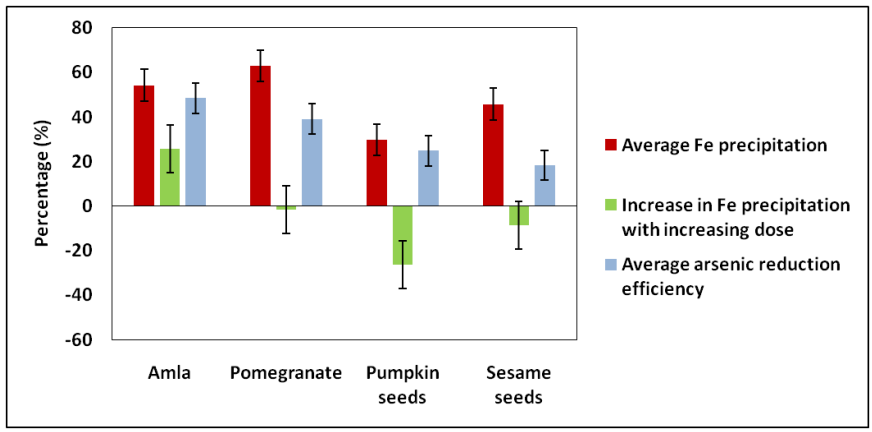


Fig. 71. Iron and arsenic co-precipitation

At the two doses, the average co-precipitation percentage of iron is observed highest in pomegranate (62.9%), followed by amla (54.2%), sesame seeds (45.8%) and pumpkin seeds (29.8%). However, with increasing dose of the components, the iron co-precipitation percentage is only increased for amla (25.7%). For the other three components, the iron co-precipitation percentage decreased with increasing doses. The decreasing percentages were 1.52%, 26.2% and 8.52 % for pomegranate, pumpkin and sesame seeds respectively. Therefore, it can be said that the leaching capacity of iron is best for amla. This is very much supportive with the previous observation, i.e. the As removal efficiency is also highest for amla (48.5%).

5.6.5.5. pH of treated drinking water

SORAS is an easy method that uses sunlight in PET- or UV-A transparent containers to decreaseAsconcentration in drinking water. Groundwater in Bengal delta contains natural Fe(II) and Fe(III) and thus SORAS could cutAs and would be available with minimal cost. It can treat small quantities of drinking water domestically. Also, closed PET are advantageousthan open bottles, because of no chance of contamination carried by the wind, least loss of CO_2that increasesthe pH, precipitation of iron(hydr)oxides and decantation of the supernatant water is possible easily when the bottles are kept vertically. Wegelin and Sommer (1998) showed that PET bottles have been used effectively for solar disinfection as they are available locally and can be reused.

The present study has been moderately successful to reduce As concentration from small amount of contaminated water. This much efficiency of SORAS is obtained without having initial iron concentration in water; only through the natural iron content of the organic materials added. The citric acid amount would catalyze the photochemical reaction. The pH of the experimental water at 2h and 4h of irradiation remains between 5.9 and 6.5. The spiked tap water was clear and colorless initially and then turned opaque within 45 min, because of oxidation of Fe(II) to Fe(III) and formation of Fe(III)(hydr)oxide colloids. The rate of Fe(II) oxidation is intensely pH-dependent. At pH 7.0 Fe(II) is oxidized within 15-30 min and at pH 8.0 within 9-15 sec (Stumm and Lee 1961).

5.6.5.6. Cost measurement

A market survey has also been done to evaluate the availability of the potential materials and their price range throughout the year. The data obtained are given in the **Table 66**.

Table 66: Cost measurement of different sources based on market survey

Ingredients	Arsenic removal percentage	Price range/100g (INR)	Availability	Comparison of price range/100g with amla (%)	Comparison of arsenic removal percentage with amla
Amla	48.5 %	6-8	Throughout the year	NA	NA
Pomegranate	39 %	15-18	Throughout the year	57.5 % more expensive	19.6 % less
Pumpkin seed	24.9 %	25-30	Throughout the year	74.5 % more expensive	`48.6 % less
Sesame seeds	18.3 %	30-50	Throughout the year	82.5 % more expensive	62.2 % less

It can be said that pumpkin seeds and sesame seeds As removal efficiency is not significant as well as they are more expensive than amla. Amla or Indian gooseberry is apparently the most inexpensive component among the four studied materials. Its price at 100 g is approximately 57.5% less than pomegranate, 74.5% less than pumpkin seeds and 82.5% less than sesame seeds. Most importantly, As removal efficiency of amla juice is also found to be highest (48.5%) in the present study when no external iron salt has been applied in the water or else initial iron concentration of the water is very low. Removal performance of As by amla is higher than pomegranatae by 19.6%, pumpkin seeds by 48.6 % and sesame seeds by nearly 62.2%. Arsenic removal efficiency of the SORAS method by Wegelin et al. (2000) using small citric acid doses is observed between45 and 78% (average 67 %) in absence of externally added iron salt. The initial iron concentration however was about 5 mg/L. They reported that SORAS can treat raw water containing an Asconcentrationbelow 100 – 150 mg/L provided sufficient iron and UV-A intensity is available. Amla treated water was slightly sour in the taste but can be easily used for cooking and drinking.

Iron content and vitamin-Cenriched components can initiate reduction of As even when the indigenous iron concentration of the water sample is low. Amla has showed highest As reduction percentage among the four studied materials. Although, the rate of co-precipitation of Aswith iron was slow compared to other studies as any kind of inorganic iron salts were not applied directly. The As removal efficiency of this experiment is less as compared to other studies that have been done with inorganic iron salts, its efficiency exclusively in household scenario is highly considerable. Amla is found to be most

efficient and easily available as well as cost-effective ingredient to promote removal of As using solar oxidation. The average As removal efficiency of amla and pomegranate are 48.5% and 39% respectively. Arsenic reduction efficiency of amla was better at dose I (49.3%) compared to dose II (47.8%) whereas the same for pomegranate was opposite. Efficiency of As reduction for pomegranate was higher at dose II (39.4%) than dose I (38.6%). Amla is available in the market throughout the year. Also, it is the most cost efficient organic component in the study. Application of solar oxidation is energy efficient and does not consider any external force. Considering, the present scenario of As contamination in West Bengal, this procedure is beneficial in household level to control scarcity of pure drinking water and remediation of Asfrom the same. Doses of amla juice and time of irradiation can be optimized more in future to achieve better removal efficiency. This work can be extended in future with varying As and iron concentration levels.

Chapter 6:

Conclusion

Consumption and usage of raw groundwater in the studied sites of West Bengal is in a doubtful situation. After several mitigation strategies undertaken, the quality of drinking water in the studied area is still abysmal and the population confronts a high health risk from prolonged consumption of contaminated water. The groundwater quality of Nadia is hard and tends to be alkaline apart from being toxic with reference to presence of As in drinking water in all the 17 blocks. Numerous As patients are there struggling to survive in the villages of Nonaghata, Ghetugachi in the blocks of Haringhata and Chakdah, the two most affected blocks in the said district. A matter of angst lies there from the other toxic chemicals like NO_3^-; few samples in four blocks have been recognised with surplus concentration (45 to 57.6) mg/L. Groundwater quality index reveals that 38.2% and 19.1% water are categorized as 'poor' and 'very poor' while 9.1% water samples are exceptionally 'inappropriate for drinking'. The concentration of U rangesunder its allowable limit but it is essential to point out that U dissolution in groundwater may aggravate with the concentrations of Cl^-, NO_3^- and HCO_3^-, carrier of U. Fluoride initiated contamination is not detected yet, however, there happens amajor cancer risk from As and U and also anoteworthy non-carcinogenic risk from NO_3^-. Assessment of groundwater quality in Bardhaman district also shows NO_3^- contamination apart from As contamination in two blocks. Special attention should be given on excessive use of NO_3^- fertilzers in agricultural fields as high NO_3^- in groundwater can cause serious health problems in children like blue baby syndrome or infant cyanosis. The domestic and community tube-well and government supplied pipeline service water in Raninagar II, Murshidabad are found to be unsafe for drinking irrespective of Fe/As ratios 33.0 and 36.6. The government has supplied river treated water through pipeline for drinking purpose which is not either available consistently or the water is not originally surface treated. There exists a gap between water supply system and community welfare. Sometimes, the pipelines are tapped unlawfully which creates damage. Arsenic removal plants are also not well maintained. The negligence and lack of awareness signify that both the affected population and the administration underestimate the benefits of As remediation as it is invisible and odorless n water and it affects people with time. R.O. plants and Sajaldhara water treatment plants had respective As removal efficiency of 77.6 and 74.4 %, yet they need regular monitoring. Among all the available drinking water sources, dug wells are apparently the safest option with a Fe/As ratio of 66.1, showing lowest health risk. In North 24 Parganas, the picture of arsenic removal plants (ARP) is also quite distressing; average annual As removal

efficiency of the ARPs are 61.2% (range: 35.2 to 82.6%). Even due to ill maintenance two ARPs got closed during my study period. 25% treated water samples are found with 'poor' quality and 16.7% treated water samples are of 'high' heavy metal evaluation index value which signifies that the inhabitants consume contaminated water unknowingly. In addition, food chain As-contamination triggers a daily threat as approximately 12.5 tonnes of As is withdrawn from groundwater each year for irrigation and deposited on irrigated soil (1.08 kg/ha). The concentrations of beneficial micronutrients in groundwater are low and their inadequate intake is the probable explanation behind the increased As toxicity in the studied area. Arsenic contamination has spread into both the conventional drinking water and the substitute drinking water sources, which manifest carelessness as well as the incognizance of the administrations. About, 45.5 and 60 % of drinking water samples from Sajaldhara water treatment plants and pipeline supply are As-contaminated above the acceptable limit in drinking water. Conclusion comes from this research that the biomarkers As is related to age of the individuals, while independent of sex. In each age group, As concentration in nail is greater than hair by 3.13 times approximately. It is statistically proved that nail As concentration is the most efficient biomarker for determination of As induced toxicity in body system. The adults suffer from an advanced order of health risk compared to the other two age groups. Tube-well water gives highest health risk in comparison to pipeline supplied water or Sajaldhara plant treated water. The estimated mean cancer risk value from the other drinking water sources is undeniably greater than the acceptable risk. Sensitivity analysis explains that the most imperativeparameterregulating health risk of the populace is 'As concentration' in drinking water and rice grain compared to 'exposure duration' and 'body weight'. Hence, an alteration in daily diet can trim down the health risk. Besides human, drinking water As concentration from the exposed area beats the optimum limit of As for the livestock. Paddy straw and rice husk contain an elevated level of As, which makes it exceed the maximum level of As in animal feed according to European Union. In exposed cattle population, the per capita ingestion of As per day is ~4.56 times higher than control. In exposed cattle and goat population, the maximum consumed As through drinking water is excreted fast hrough urine, whereas, As from paddy straw is excreted more through urine than faeces. Arsenic exposure in the domestic livestock produces sub-clinical toxicity along with health risk inhumans through animal sourced products. However, As concentration in milk didn't cross the permissible limit but the estimated health risk from it is not insignificant. The casein part entraps maximum As due to the active presence of phosphoserine. Among the

animal proteins, consumption of whole cow milk causes higher risk than egg or meat. Nevertheless, in the endemic sites, contaminated drinking water and rice gives utmost risk. The high accretion of As in animal excretion makes the soil infertile, drops the quality of crops. This cycle fosters bio-magnification as the metal goes into next trophic layers; plant, grains, animal systems and finally humans. Following all the issues, a solar oxidation process is promoted in my research at domestic level for removal of As (removal efficiency~50%) from contaminated drinking water with application of amla at minor dose.

Arsenic contamination in 2022 is not limited only in groundwater but has spread in every trophic level of our ecosystem. Therefore, for an As-free healthy environment, community participation and administrative intervention should go hand in hand. The major suggestions for the affected population are:

- Usage of dug well (both at personal and community level), conservation of surface water bodies, as well as, rainwater harvesting.
- Domestic livestock should also be fed with suface water; their excreta and the excess agrarian wastes should be managed scientifically and carefully.
- Active use of solar radiation method in contaminated drinking water with addition of small doses of amla juices.
- Intensification of monsoonal paddy cultivation for reduced use of groundwater.
- Practise of paddy cultivation by use of drip irrigation or sprinkler irrigation method.
- Extensive cultivation of low As accruing rice cultivars.
- Consumption of vit-C, vit E, selenium and zinc enriched nutritious food beside As-free drinking water.

A regular monitoring of the pipeline water quality before to reach to the affected mass is the prime call of the hour which should be addressed by the administrations.

I hope that this entire research would draw the attention of the policy makers to make a restriction on ‘groundwater exploitation’ and be helpful for the society.

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Future scope of my research

- Evaluation of irrigational water quality in several As-exposed sites of West Bengal for selective staple crops like paddy, wheat etc in comparison to As-unexposed sites.
- Finding a threshold value of bio-accumulation factor of arsenic in rice grain from contaminated water and soil both in arsenic exposed and unexposed sites of Bengal.
- Comparative evaluation of the alternate paddy cultivation practices throughout India for reducing arsenic accumulation in rice.
- A hypothesis-based investigation on the significance of the prime factors regulating arsenic toxicity and associated health risk in a arsenic unexposed population compared to an exposed population of West Bengal, India.
- Evaluation of the range of bio-magnification of arsenic in the subsequent trophic levels of our ecosystem.
- Adverse impacts of arsenic on the nutritional content of the consumable animal by products from arsenic exposed livestock compared to arsenic unexposed livestock produced items.

Publications

Full Articles (Papers):

1st authorship:

- **Das, A.,** Joardar, M., Chowdhury, N. R., Mridha, D., De, A., Majumder, S., Das, J., Majumdar, K. K., Roychowdhury, T. (2022). Significance of the prime factors regulating arsenic toxicity and associated health risk: a hypothesis-based investigation in a critically exposed population of West Bengal, India. ENVIRONMENTAL GEOCHEMISTRY AND HEALTH. Nov 6:1-24.

- **Das, A.,** Joardar, M., De, A., Mridha, D., Chowdhury, N.R., Khan, M.T.B.K., Chakrabartty, P. and Roychowdhury, T. (2021). Pollution index and health risk assessment of arsenic through different groundwater sources and its load on soil paddy-rice system in a part of Murshidabad district of West Bengal, India. GROUNDWATER FOR SUSTAINABLE DEVELOPMENT, 15,100652. https://doi.org/10.1016/j.gsd.2021.100652

- **Das, A**., Joardar, M., Chowdhury, N. R., De, A., Mridha, D., & Roychowdhury, T. (2021). Arsenic toxicity in livestock growing in arsenic endemic and control sites of West Bengal: risk for human and environment. ENVIRONMENTAL GEOCHEMISTRY AND HEALTH, 43(8), 3005-3025. https://doi.org/10.1007/s10653-021-00808-2

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Co-authorship:

- Nath, B., **Das, A**., Majumder, S., Roychowdhury, T., Ni-Meister, W. & Rahman, M. M. (2022). Geospatial Machine Learning Prediction of Arsenic Distribution in

the Groundwater of Murshidabad District, West Bengal, India: Analyzing Spatiotemporal Patterns to Understand Human Health Risk. ACS ES&T WATER. 10.1021/acsestwater.2c00263

- Joardar, M., **Das, A.,** Chowdhury, N. R., Mridha, D., Das, J., De, A., Majumder, S., Majumdar, K. K. & Roychowdhury, T. (2022). Impact of treated drinking water on arsenicosis patients with continuous consumption of contaminated dietary foodstuffs: A longitudinal health effect study from arsenic prone area, West Bengal, India. GROUNDWATER FOR SUSTAINABLE DEVELOPMENT, 100786. https://doi.org/10.1016/j.gsd.2022.100786

- De, A., Mridha, D., Joardar, M., **Das, A**., Chowdhury, N. R., & Roychowdhury, T. (2022). Distribution, prevalence and health risk assessment of fluoride and arsenic in groundwater from lower Gangetic plain in West Bengal, India. GROUNDWATER FOR SUSTAINABLE DEVELOPMENT, 100722

- Mridha, D., Ray, I., Sarkar, J., A., De, A., Joardar, M., **Das, A.,** Chowdhury, N.R., Acharya, K. and Roychowdhury, T. (2022). Effect of Sulfate Application on Inhibition of Arsenic Bioaccumulation in Rice (Oryza sativa L.) with Consequent Health Risk Assessment of Cooked Rice Arsenic on Human: A Pot to Plate Study. ENVIRONMENTAL POLLUTION, 293, 118561.

- Ray, I., Mridha, D., Sarkar, J., Joardar, M., **Das, A.,** Chowdhury, N. R.,De, A., Acharya, K. and & Roychowdhury, T. (2022). Application of potassium humate to reduce arsenic bioavailability and toxicity in rice plants (Oryza sativa L.) during its course of germination and seedling growth. ENVIRONMENTAL POLLUTION, 120066.

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Through Dietary Intakes: A Village Level Study from West Bengal, India. IN CONFERENCE GEOLOGICAL SOCIETY OF INDIA (178-184).

- Joardar, M., **Das, A.,** Chowdhury, N. R., Mridha, D., De, A., Majumdar, K. K., & Roychowdhury, T. (2021). Health effect and risk assessment of the populations exposed to different arsenic levels in drinking water and foodstuffs from four villages in arsenic endemic Gaighata block, West Bengal, India. ENVIRONMENTAL GEOCHEMISTRY AND HEALTH, 3(8), 3027-3053.

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- Mridha, D., Paul, I., De, A., Ray, I., **Das, A.,** Joardar, M., Chowdhury, N.R., Bhadoria, P.B.S. and Roychowdhury, T. (2021). Rice seed (IR64) priming with potassium humate for improvement of seed germination, seedling growth and antioxidant defense system under arsenic stress. ECOTOXICOLOGY AND ENVIRONMENTAL SAFETY, 219,112313.

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- Chowdhury, N.R., **Das, A.,** Mukherjee, M., Swain, S., Joardar, M., De, A., Mridha, D. and Roychowdhury, T. (2020). Monsoonal paddy cultivation with phase-wise arsenic distribution in exposed and control sites of West Bengal, alongside its assimilation in rice grain. JOURNAL OF HAZARDOUS MATERIALS, 400, 123206.

- Chowdhury, N.R., **Das, A.,** Joardar, M., De, A., Mridha, D., Das, R., Rahman, M.M. and Roychowdhury, T. (2020). Flow of arsenic between rice grain and water: Its interaction, accumulation and distribution in different fractions of cooked rice. SCIENCE OF THE TOTAL ENVIRONMENT, 731, 138937.

- Bhattacharya, P., Adhikari, S., Samal, A. C., Das, R., Dey, D., Deb, A., Ahmed, S., Hussein, J., De, A., **Das, A.,** Joardar, M., Panigrahi, A. K., Roychowdhury, T. &Santra, S. C. (2020). Health risk assessment of co-occurrence of toxic fluoride and arsenic in groundwater of Dharmanagar region, North Tripura (India). GROUNDWATER FOR SUSTAINABLE DEVELOPMENT, 11, 100430.

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Book Chapter:

1st authorship:

- **Das, A.,** Chowdhury, N. R., Joardar, M., Mridha, D., De, A., Majumder, S. &Roychowdhury, T. Food Chain Arsenic Contamination in West Bengal, India: Accumulation, Distribution and Human Health Risk. IN ENVIRONMENTAL CONTAMINANTS IMPACT, ASSESSMENT, AND REMEDIATION, edited by Ganguly, P., Mandal, J., Paramsivam, M. & Patra, S. Apple Academic Press (In press).

- **Das, A.,** Chowdhury, N. R., Mridha, D., Joardar, M., De, A., Majumder, S. & Roychowdhury, T. (2022). Accumulation and Translocation of Arsenic in Rice with its Distributional Flow during Cooking: A Study in West Bengal, India. IN ENVIRONMENTAL SCIENCE AND ENGINEERING: GLOBAL ARSENIC

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Co-authorship:

- Chowdhury, N. R., **Das, A.,** Joardar, M., Mridha, D., De, A., Majumder, S., Mandal, J., majumdar, A. & Roychowdhury, T (2022). Distribution of Arsenic in Rice Grain from West Bengal, India: Its Relevance to Geographical Origin, Variety, Cultivars and Cultivation Season. IN ENVIRONMENTAL SCIENCE AND ENGINEERING: GLOBAL ARSENIC HAZARD ECOTOXICOLOGY AND REMEDIATION, edited by Niazi, N. K., Bibi, I. & Aftab, T. (pp. 509-531), Springer.
- Chowdhury, N. R., Sinha, D., **Das, A.,** Joardar, M., Joseph, A., Ray, I., Mridha, D., De, A. & Roychowdhury, T. (2022). A Potential Phytoremedial Strategy for Arsenic from Contaminated Water Using *Hygrophila spinosa* (Starthorn leaves). IN ARSENIC IN PLANTS: UPTAKE, CONSEQUENCES, AND REMEDIATION TECHNIQUES, edited by Srivastava, P. K., Singh, R., Parihar, P. & Prasad, S. M. Wiley
- Das, A., **Das, A.,** Mukherjee, M., Das, B., Mukherjee, S. C., Pati, S., & Roychowdhury, T. (2020). Status of Groundwater Arsenic Contamination in the GMB Plain. IN MANAGING WATER RESOURCES AND HYDROLOGICAL SYSTEMS (pp. 369-381). CRC Press.
- Chowdhury, N. R., Joardar, M., **Das, A.,**& Roychowdhury, T (2021). Evaluation of Arsenic Entry Routes into Rice Grain during Harvesting, Post-Harvesting of Paddy and Cooked Rice Preparation. IN MULTIDIMENSIONAL APPROACHES TO IMPACTS OF CHANGING ENVIRONMENT ON HUMAN HEALTH (pp. 219-245). CRC Press.

- Joardar, M., Chowdhury, N. R., **Das, A.,**& Roychowdhury, T. (2021). Evaluation of Health Effects and Risk Assessment of Arsenic on an Unexposed Population from an Arsenic-Exposed Zone of West Bengal, India. IN MULTIDIMENSIONAL APPROACHES TO IMPACTS OF CHANGING ENVIRONMENT ON HUMAN HEALTH (pp. 113-134). CRC Press.
- Chowdhury, N.R., Joardar, M., **Das, A**., Joseph, A., Sinha, D., Chakraborti, R., Kar, D. and Roychowdhury, T. (2021). Arsenic Modulates Health Hazards Through Dietary Intakes: A Village Level Study from West Bengal, India. IN CONFERENCE GEOLOGICAL SOCIETY OF INDIA (178-184).

Presentations

or

Conference proceedings

- **Presented** a technical paper on "Evaluation of groundwater quality in some arsenic exposed sites of West Bengal, India and implemented mitigation strategies" in the *Web-based Exposition on Engineering and Technology Research at Jadavpur University, WEBINAR-FET-JU R&D Expo 2021*, organized by **R&D Committee, TEQIP-III, Jadavpur University** during February 26-27, 2021.
- **Presented** a paper on "Ground Water Quality Assessment and Health Risk Evaluation in Nadia, West Bengal, India: An Alertness During Pandemic" in the *International Interdisciplinary Conference* on "*COVID 19: Challenges and Impact on Health, Environment, Livelihood and Education"* organized by the **Scientific and Environmental Research Institute, Kolkata,** held on 28 - 30 May, 2021.
- **Presented** a paper on "Role of arsenic contaminated water during cooking of rice grain: an inverse relation" in the *8th International Congress and Exhibition on Arsenic in the Environment, Bridging Science to Practice for Sustainable Development (As2021),* organized digitally at the **Wageningen University and Research, Wageningen, The Netherlands** from 7-9 June, 2021.
- "Assessment of groundwater quality in an arsenic-affected district, Nadia of West Bengal with special reference to radioactive uranium."**A. Das,** S. S. Das, M. Joardar, N. Roy Chowdhury, S. Swain, A. De, T. Roychowdhury. *Proceedings in Twentieth National Symposium on Environment (NSE-20), Focal Theme: Challenges in energy resource management & climate change.* Eds. R. M. Tripathi, M. Kumar, S. K. Jha, V. Jain, A. v. Kumar, V. Pulhani, I. V. Saradhi, A. C. Patra, M. K. Mishra, S. K. Sahoo. 2020. **Health, Safety & Environment Group, Bhabha Atomic Research Centre, Mumbai and Indian Institute of Technology, Gandhinagar, Board of Research in Nuclear Sciences, Department of Atomic Energy**, Govt. of India, pp. 95-96.
- Groundwater arsenic contamination with special reference to its entry in rice grain post-harvest in Bengal delta. T. Roychowdhury, N. Roy Chowdhury, M. Joardar, S. Swain, **A. Das**, M. Mukherjee, A. De, S. Ghosh, D. Saha, B. Ghosh, S. S. Das, S. Majumder. *Proceedings inTwentieth National Symposium on Environment (NSE-20), Focal Theme: Challenges in energy resource management & climate change.* Eds. R. M. Tripathi, M. Kumar, S. K. Jha, V. Jain, A. v. Kumar, V. Pulhani, I. V. Saradhi, A. C. Patra, M. K. Mishra, S. K. Sahoo. 2020. **Health, Safety &**

Environment Group, Bhabha Atomic Research Centre, Mumbai and Indian Institute of Technology, Gandhinagar, Board of Research in Nuclear Sciences, Department of Atomic Energy, Govt. of India, pp. 101-102.

- Arsenic toxicity through drinking water and rice grain with special reference to health effects: A village level study from West Bengal, India. N. Roy Chowdhury, M. Joardar, S. Swain, **A. Das**, A. De, M. Mukherjee, T. Roychowdhury.*Proceedings in Twentieth National Symposium on Environment (NSE-20), Focal Theme: Challenges in energy resource management & climate change.* Eds. R. M. Tripathi, M. Kumar, S. K. Jha, V. Jain, A. v. Kumar, V. Pulhani, I. V. Saradhi, A. C. Patra, M. K. Mishra, S. K. Sahoo. 2020. **Health, Safety & Environment Group, Bhabha Atomic Research Centre, Mumbai and Indian Institute of Technology, Gandhinagar, Board of Research in Nuclear Sciences, Department of Atomic Energy, Govt. of India**, pp. 309-310.
- **Presented** paper titled "Assessment of Groundwater quality of Nadia, West Bengal: Health risk appraisal for drinking and household purpose" in the *International Conference on Sustainable Development & Climate Change*, organized by **Department of Environmental Science, Amity University Madhya Pradesh, Gwalior, India** during February 10-11, 2020.
- "Arsenic changing pattern in paddy plants throughout the pre monsoon cultivation period and its further entry in parboiled rice grain during post harvesting technology". N. Roy Chowdhury, M. Joardar, S. Swain, **A. Das**, A. De, S. Majumder, T. Roychowdhury. *National Seminar on 'Groundwater arsenic contamination problem in Ganga-Meghna-Brahmaputra (GMB) plain: Its health effects, socio-economic implications and mitigation strategies."***Department of Economics, VijaygarhJyotish Ray College, Jadavpur in collaboration with School of Environmental Studies, Jadavpur University, VijaygarhJyotish Ray College,** 25th March, 2019.
- "Arsenic accumulation in food chain of West Bengal, India: Special reference to rice grain and its potential health hazard to population". S. Swain, N. Roy Chowdhury, M. Joardar, **A. Das**, M. Mukherjee, T. Roychowdhury. National Seminar on '*Groundwater arsenic contamination problem in Ganga-Meghna-Brahmaputra (GMB) plain: Its health effects, socio-economic implications and*

mitigation strategies."**Department of Economics, VijaygarhJyotish Ray College, Jadavpur in collaboration with School of Environmental Studies, Jadavpur University, VijaygarhJyotish Ray College**, 25th March, 2019.

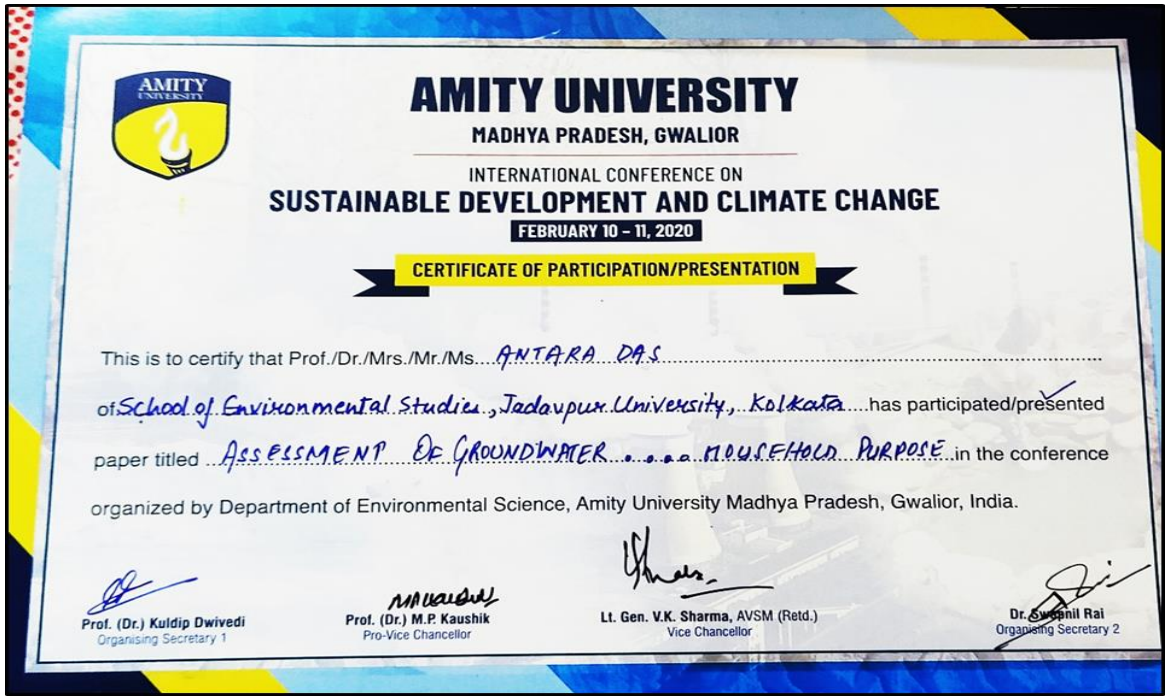
- Arsenic toxicity through drinking water and rice grain with special reference to health effect: a school children study from arsenic affected area in West Bengal, India. M. Joardar, N. Roy Chowdhury, S. Swain, **A. Das,** A. De, S. Majumder, T. Roychowdhury. "*National Seminar on 'Groundwater arsenic contamination problem in Ganga-Meghna-Brahmaputra (GMB) plain: Its health effects, socio-economic implications and mitigation strategies*". **Department of Economics, VijaygarhJyotish Ray College, Jadavpur in collaboration with School of Environmental Studies, Jadavpur University, VijaygarhJyotish Ray College**, 25th March, 2019.

- An insight of arsenic contamination in groundwater and food chain with special reference to health effects on domestic animals. **A. Das**, M. Joardar, N. Roy Chowdhury, S. Swain, A. De, M. Mukherjee, B. Ghosh, S. Das, S. Majumder, T. Roychowdhury. *"National Seminar on 'Groundwater arsenic contamination problem in Ganga-Meghna-Brahmaputra (GMB) plain: Its health effects, socio-economic implications and mitigation strategies".* **Department of Economics, VijaygarhJyotish Ray College, Jadavpur in collaboration with School of Environmental Studies, Jadavpur University, VijaygarhJyotish Ray College**, 25th March, 2019.

- **Participated** inthe *National Conclave on Water Resources Management*, jointly organized by **Academy of Water Technology & Environ Management & CSIR-Central glass & Ceramic Research Institute, Jadavpur** on January 17-18, 2019.

- **Presented** paper titled "Arsenic accumulation in paddy plant during pre-monsoon cultivation and its additional entry in rice grain through Post harvesting technology", **Das, A.,** Chowdhury, N. R., Joardar, M., De, A., Mridha, D., Ghosh, R., Bhowmick, A., Roychowdhury, T. in 1stInternational *Conference on Energy Management for Green Environment (UEMGREEN) & biodiversity conservation ecosystem managemen)*, 2019, pp. 1-8, doi: 10.1109/UEMGREEN46813.2019.9221381.

- "Groundwater arsenic contamination with special reference to food chain in West Bengal, India: Impact, Measurement and Remediation Strategies" Madhurima Joardar, Nilanjana Roy Chowdhury, Shrestha Swain, **Antara Das**, Anuja Joseph, Debapriya Sinha, SouravMaity, Tarit Roychowdhury at *national seminar on Environmental Challenges: Monitoring, Assessment and Remediation* organized by **Department of Zoology, Patna University, Patna (Bihar)**, 2018.
- "Groundwater arsenic contamination with special reference to food chain in West Bengal, India: Magnitude, Health effects and Remedial Strategies" Nilanjana Roy Chowdhury, Madhurima Joardar, **Antara Das**, Shrestha Swain, Anuja Joseph, Sourav Maity, Debapriya Sinha, Duhita Kar, Rishika Chakraborti, Tarit Roychowdhury at *Brain Storming Session on Water Resources of Eastern (West Bengal, Bihar, Jharkhand) and North Eastern States of India* organized by **Centre for Ground Water Studies in association with Geological Society of India**, Kolkata, 2018.
- Participated in **5th NUP workshop** on "**Statistical Analysis of Data Generated under National Uranium Project"** held at **BARC, Mumbai** during September 26-27, 2018.



AMITY UNIVERSITY

MADHYA PRADESH, GWALIOR

INTERNATIONAL CONFERENCE ON

SUSTAINABLE DEVELOPMENT AND CLIMATE CHANGE

FEBRUARY 10 – 11, 2020

CERTIFICATE OF PARTICIPATION/PRESENTATION

This is to certify that Prof./Dr./Mrs./Mr./Ms. ANTARA DAS

of School of Environmental Studies, Jadavpur University, Kolkata has participated/presented paper titled ASSESSMENT OF GROUNDWATER ... HOUSEHOLD PURPOSE in the conference organized by Department of Environmental Science, Amity University Madhya Pradesh, Gwalior, India.

Prof. (Dr.) Kuldip Dwivedi
Organising Secretary 1

Prof. (Dr.) M.P. Kaushik
Pro-Vice Chancellor

Lt. Gen. V.K. Sharma, AVSM (Retd.)
Vice Chancellor

Dr. Sushil Rai
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THE 8TH INTERNATIONAL

CONGRESS & EXHIBITION ON ARSENIC IN THE ENVIRONMENT

7th – 9th June 2021 | The Netherlands

As

Participation Certificate

This is to certify that

Antara Das

Has participated as oral speaker in the 8th International Congress and Exhibition on Arsenic in the Environment, Bridging Science to Practice for Sustainable Development (As2021) organized digitally at the Wageningen University and Research, Wageningen, The Netherlands from 7-9 June, 2021.

Prof. Dr. Albert van der Wal
KWR Water Research Institute Nieuwegein, The Netherlands

Dr. Arslan Ahmad
Wageningen University and Research, Wageningen, The Netherlands

Prof. Dr. Prosun Bhattacharya
KTH Royal Institute of Technology Stockholm, Sweden

Memories

Working images in lab, field and at BARC, Mumbai during NUP workshop on "Statistical Analysis of Data Generated under National Uranium Project" dated 26th-27th September, 2018 & at AMITY University, Gwalior during "International Conference on Sustainable Development & Climate Change" organized by Department of Environmental Science dated 10th-12th February, 2020

Field images at villages of Gaighata, North 24 Parganas accompanied by Dr. Kunal Kanti Majumdar (KPC Medical College); dated: 8th March, 2020

Field images at villages of Gaighata, North 24 Parganas; dated: 3rd August, 2019 & 12th September, 2021

Media coverage of my work

(Newspapers & magazines)

এই সময়

Dated: 24th February, 2020; by Koushik Sarkar

নদিয়ার চার ব্লকে নাইট্রেট-বিপদ

যাদবপুরের গবেষণায় সামনে এল তথ্য, আরও ৫ জেলায় সমীক্ষা

কৌশিক সরকার

নাইট্রেটের প্রভাব

- মস্তিষ্ক বিকিয়ে মেলোজ, মাথা ঝিমঝিম করা
- দুতদুত শ্বাস কষ্ট
- পেটি পেটের সমস্যা
- ব্লক অ্যালার্জি, এরিথেমা, ইডিমা
- চুল চুল পড়ে যায়
- নখ সায়ানোসিসের কারণে নখ নীল হয়ে যায়

বর্তমান

Dated: 10th August, 2021; by Abhisek Pal

কৃষিতে ব্যবহৃত রাসায়নিকই ভিলেন ভূগর্ভস্থ জলে বিষের সন্ধান, উদ্বেগ

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Dated: 12nd March, 2021; by Snigdhendu Bhattacharya

BENGAL/ARSENIC CONTAMINATION

A Can Of Poison

Bengal's arsenic challenge grows bigger as the toxin seeps into the food chain—grain, greens, dairy, meat

Outlook

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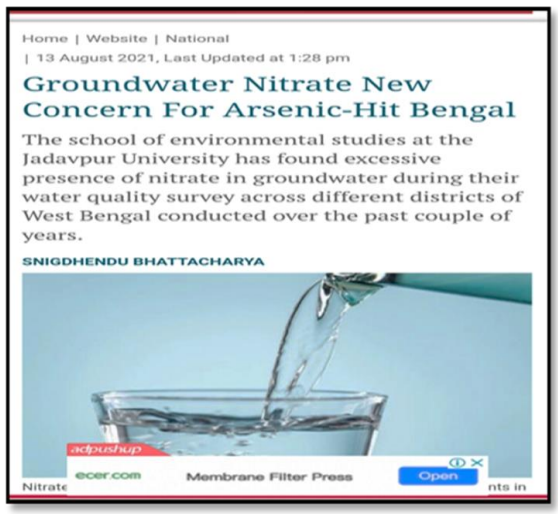

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| 13 August 2021, Last Updated at 1:28 pm

Groundwater Nitrate New Concern For Arsenic-Hit Bengal

The school of environmental studies at the Jadavpur University has found excessive presence of nitrate in groundwater during their water quality survey across different districts of West Bengal conducted over the past couple of years.

SNIGDHENDU BHATTACHARYA

বর্তমান

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দুধ সহ সব প্রাণিজ খাদ্যদ্রব্যে
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DownToEarth 25 years
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Dated: 10th September, 2021; by Umesh Kumar Roy

बंगाल: मवेशियों के दूध, अंडे और गोश्त में भी मिले आर्सेनिक

उबले हुए अंडे के सफेद हिस्से में आर्सेनिक की मौजूदगी 9.7-30.6 माइक्रोग्राम प्रति किलोग्राम मिली और पीले हिस्से में 9.78-22.1 माइक्रोग्राम प्रति किलो आर्सेनिक मौजूद था।

By Umesh Kumar Roy
On: Friday 10 September 2021

अगली खबर ›

Stades
Stories that inspire

Dated: 8th November, 2021; by Partho Barman

ENVIRONMENT, SOCIETY

West Bengal's Gaighata: Where arsenic toxicity in livestock is now adding to poison in human bodies

POSTED ON NOVEMBER 8, 2021 BY PARTHO BURMAN

08 Nov

Biswajit Das, a 55-year-old farmer from Sutia (also Shutia) village in West Bengal's Gaighata block, North 24 Parganas district, has discontinued farming. No, it's not because his land has turned infertile or he is incurring losses. It is because Biswajit has developed chronic bronchitis and lost the physical vitality to even perform daily chores.

INDIA TODAY

Dated: 28th November, 2022; by Romita Datta

UP FRONT

WEST BENGAL

BEWARE OF THAT MAACHH-BHAAT

By Romita Datta

In Bengal, they say even the most exacting of palates can be satiated with a hearty meal of maachh-bhaat, or fish and rice. However, in a pincer attack of sorts, both face a threat of contamination by chemical poisoning. While arsenic in its inorganic form, occurring in soil and groundwater, is the blight that is poisoning varieties of paddy, mercury in water and the use of formalin are the villains infecting fish. Alarms raised in either case have failed to elicit an appropriate response.

It's known since the 1980s that large swathes of West Bengal fall in the naturally arsenic-contaminated Ganga-Brahmaputra-Meghna basin. Groundwater infected by arsenic and its consumption has been a major health concern for long. The entry of arsenic in the food chain through its presence in paddy grain has been established in research by scientists of the School of Environmental Studies (SOES) in Jadavpur University for the past several years. Dr Tarit Roychowdhury, senior faculty member of the school and water expert, who has studied arsenic contamination in soil and water and its effects on crops and vegetables, found out that eight rice-producing districts—North and South 24 Parganas, Nadia, Howrah, Hooghly, Bardhaman, Malda and Murshidabad—lie in the state's arsenic belt. Arsenic is listed as Group 1 carcinogen by the International Agency for Research on Cancer. It's linked to cancers of the skin, liver and kidney and can cause miscarriages and stillbirths. Crops contaminated with arsenic can, in the most alarming projection, slow-poison a mass of human population. Arsenic as an inorganic chemical is found in groundwater at a shallow depth of 40 metres. Most shallow tube-wells of Bengal are between 20 feet and 100 feet.

Crop-contamination happens when arsenic-laden groundwater is used for irrigation. Thus, pre-monsoon paddy cultivated with groundwater runs a high risk of getting polluted with arsenic. According to agricultural research, rice paddy can absorb arsenic 10 times more than wheat and barley, because its roots are required to stand in flooded fields for a considerable period, resulting in maximum absorption of arsenic. While studying the progress of arsenic from root to shoot, Roychowdhury has observed that while its mobility is highest at the roots, other parts of the plant are also susceptible to the carcinogenic effects of the chemical. Studies have revealed that the edible rice grain has the least amount of arsenic accumulation in comparison to leaves and the shoot, but still contains 50-1000 ppb (parts per billion) of arsenic, whereas the prescribed limit of inorganic arsenic by the European Commission (2015) and Codex Alimentarius Commission (2014) is 100-1000 ppb depending on different geo-environmental conditions.

MOSTLY...BUT SAFE? The beloved rice and fish curry of Bengalis could be laced with poison

This is not all. Paddy shoots, used as feed for livestock, thus contain a high percentage of arsenic, putting at risk the animals and humans who consume the meat/ milk of cows and goats. According to Roychowdhury, a study on livestock, mainly cows, has revealed that cow milk, dung, urine and tail hair have traces of arsenic. While infected goat meat contains an arsenic concentration of 103-124 ppb, chicken meat contains 51 to 160 ppb. Worryingly, if arsenic present in cow urine/ dung gets mixed with the soil, it can recharge the arsenic table and start a vicious cycle of contamination, says Antara Das, senior research fellow, SOES.

22 | INDIA TODAY | NOVEMBER 28, 2022

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